ELECTRONICALLY FILED

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Non-Provisional Patent Application of: Inolex Investment Corporation			
Conf. No.:	Not Yet Assigned	Group Art Unit:	Not Yet Assigned
Appln. No.:	Not Yet Assigned	Examiner:	
Filing Date:	Herewith	Attorney Docket No.:	I0113-198U2
Title:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making The Same.		

PRELIMINARY AMENDMENT IN NEW NON-PROVISIONAL APPLICATION UNDER 37 C.F.R. 1.53(b)

Preliminary to the calculation of fees and examination of the above application being transmitted herewith, please amend the above-identified application, without prejudice, as follows:

Symrise AG, EX1006
Symrise AG v. Inolex Inc.
IPR2023-01439

Amendments to and Listing of the Claims:

Please cancel claims 1-41 without prejudice to the filing of one or more continuation or divisional applications so that the claims read as follows, with the changes as shown:

1.-41 (canceled)

42. (original) A composition, comprising at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

43. (original) The composition according to claim 42, wherein the composition is a personal care composition.

44. (original) The composition according to claim 43, wherein the personal care composition is a hair care composition, an oral care composition, a skin care composition or a cosmetic composition.

45. (original) The composition according to claim 42, wherein the composition is a composition for a household product.

46. (original) The composition according to claim 45, wherein the household product is a fabric care product or a cleaning product.

47. (original) The composition according to claim 42, wherein the composition is an industrial composition.

48. (original) The composition according to claim 42, wherein the composition is a pharmaceutical composition, a vitamin composition, or a health care composition.

49. (original) The composition according to claim 42, comprising a second bio-1,2-alkanediol, different than the first bio-1,2-alkanediol, the second bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a second bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

50. (original) The composition according to claim 49, wherein the composition further comprises at least one other bio-compound different from the first and the second bio-1,2-alkanediols.

51. (original) The composition according to claim 50, wherein the composition comprises an antimicrobial compound different from the first and the second bio-1,2-alkanediol and different from the other bio-compound.

52. (original) The composition according to claim 42, wherein the composition further comprises at least one other bio-compound different from the at least one bio-1,2-alkanediol.

53. (original) The composition according to claim 52, wherein the composition comprises at least one antimicrobial compound different from the at least one bio-1,2-alkanediol and different from the other bio-compound.

54. (original) The composition according to claim 42, wherein the first bio-1-alkene has a regioselectivity of about 95% to about 99%.

55. (original) The composition according to claim 42, wherein the first bio-alkene is bio-octene and the at least one first bio-1,2-alkane diol is bio-1,2-octanediol.

56. (original) A method for providing an antimicrobial effect to a composition, including incorporating an antimicrobial system to the composition, wherein the antimicrobial system comprises at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of at least one first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

57. (original) The method according to claim 56, wherein the antimicrobial system further comprises a second bio-1,2-alkanediol different than the first bio-1,2-alkanediol, wherein the first bio-1,2-alkanediol has a carbon chain length of about 5 to about 20 carbon atoms and is formed by conversion of a second bio-alkene, different than the first bio-alkene, having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

58. (original) The method according to claim 57, wherein the composition further comprises at least one other bio-compound different from the first and the second bio-1,2-alkanediols.

59. (original) The method according to claim 58, wherein the antimicrobial system comprises at least one other preservative compound different from the first and the second bio-1,2-alkanediol and different from the other bio-compound.

60. (original) The method according to claim 57, wherein the composition further comprises at least one other bio-compound different from the at least one first bio-1,2-alkanediol.

61. (original) The method according to claim 60, wherein the antimicrobial comprises at least one other antimicrobial compound and/or preservative compound different from the at least one first bio-1,2-alkanediol and different from the other bio-compound.

62. (original) The method according to claim 57, wherein the antimicrobial system demonstrates antimicrobial efficacy.

63. (original) The method according to claim 57, wherein the first bio-1-alkene has a regioselectivity of about 95% to about 99%.

64. (original) The method according to claim 57, wherein the first bio-alkene is bio-octene and the at least one first 1,2-alkane diol is bio-1,2-octanediol.

65. (original) A method for boosting the antimicrobial efficiency of an antimicrobial and/or a preservative in a composition, comprising incorporating an antimicrobial system into the composition, wherein the second antimicrobial system comprises at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

66. (original) The method according to claim 65, wherein the antimicrobial system further comprises a second bio-1,2-alkanediol different than the first bio-1,2-alkanediol, wherein the second bio-1,2-alkanediol has a carbon chain length of about 5 to about 20 carbon atoms and is formed by conversion of a second bio-alkene, different than the first bio-alkene, having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

67. (original) The method according to claim 66, wherein the composition further comprises at least one other bio-compound different from the antimicrobial and/or preservative and different from the first and the second bio-1,2-alkanediols.

68. (original) The method according to claim 65, wherein the composition further comprises at least one other bio-compound different from the at least one first bio-1,2-alkanediol.

69. (original) The method according to claim 65, wherein the first bio-1-alkene has a regioselectivity of about 95% to about 99%.

70. (original) The method according to claim 65, wherein the antimicrobial system demonstrates antimicrobial efficacy.

71. (original) The method according to claim 65, wherein the first bio-alkene is bio-octene and the at least one first bio-1,2-alkane diol is bio-1,2-octanediol.

72. (original) An antimicrobial product, comprising at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

73. (original) The antimicrobial product according to claim 72, further comprising a second bio-1,2-alkanediol, different than the at least one first bio-1,2-alkanediol, the second bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms and formed by conversion of a second bio-alkene, different than the first bio-alkene, having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

74. (original) The antimicrobial product according to claim 73, wherein the product further comprises at least one other bio-compound different from the at least one first and the second bio-1,2-alkanediols.

75. (original) The antimicrobial product according to claim 74, further comprising a second antimicrobial and/or a preservative different from the antimicrobial product.

76. (original) The antimicrobial product according to claim 72, wherein the product further comprises at least one other bio-compound different from the at least one first bio-1,2-alkanediol.

77. (original) The antimicrobial product according to claim 76, further comprising a second antimicrobial and/or a preservative different from the antimicrobial product.

78. (original) The antimicrobial product according to claim 72, wherein the first bio-1-alkene has a regioselectivity of about 95% to about 99%.

79. (original) The antimicrobial product according to claim 72, wherein the antimicrobial product demonstrates antimicrobial efficacy.

80. (original) The antimicrobial product according to claim 72, wherein the first bio-alkene is bio-octene and the at least one first 1,2-alkane diol is bio-1,2-octanediol.

81. (original) A product for boosting the efficacy of an antimicrobial and/or a preservative in a composition, the product comprising at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

82. (original) The product according to claim 81, further comprising a second bio-1,2-alkanediol, different than the at least one first bio-1,2-alkanediol, the second bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms and formed by conversion of a second bio-alkene, different than the first bio-alkene, having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

83. (original) The product according to claim 82, wherein the composition further comprises at least one other bio-compound different from the at least one first and the second bio-1,2-alkanediols.

84. (original) The product according to claim 81, wherein the product further comprises at least one other bio-compound different from the at least one first bio-1,2-alkanediol.

85. (original) The product according to claim 81, wherein the first bio-1-alkene has a regioselectivity of about 92% to about 99%.

86. (original) The product according to claim 82, wherein the first bio-alkene is bio-octene and the at least one first bio-1,2-alkane diol is bio-1,2-octanediol.

87. (original) The product according to claim 81, wherein the product demonstrates antimicrobial efficacy.

REMARKS

Claims 42-87 are presently pending in this application.

Claims 1-41 have been canceled without prejudice to the filing of one or more divisional or continuation applications.

No new matter has been added by these amendments, and entry thereof and an early notice of allowance are respectfully requested.

Respectfully submitted,

INOLEX INVESTMENT CORPORATION

1/30/19 By: _____
(Date)

LYNDA L. CALDERONE
Registration No. 35,837
CALDERONE BULLOCK LLC
Two Greentree Centre
9000 Lincoln Drive East, Suite 301
Marlton, NJ 08053
Telephone: 856-446-6047
Direct Dial: 845-446-6045
Facsimile: 856-267-5370
E-Mail: lcalderone@calderonebullock.com

LLC:llc

Electronic Acknowledgement Receipt

EFS ID:	35013762
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	159443
Filer:	lynda Leigh Calderone
Filer Authorized By:	
Attorney Docket Number:	I0113-198U2
Receipt Date:	30-JAN-2019
Filing Date:	
Time Stamp:	23:47:28
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Transmittal Letter	transmittal198U2.pdf	141360 b9e4e324232bef9f73d887885e829a909dfb5c68	no	3

Warnings:

Information:					
2	Application Data Sheet	ADS198U2.PDF	1351576 1112c73d4fbbc2d69a7c3e925fadc166d7817b0a	no	9
Warnings:					
Information:					
3		nonprovisional198U2.pdf	409364 29080b759655ec15307fdfe3593162afa89d7b4a	yes	50

	Multipart Description/PDF files in .zip description		
	Document Description	**Start**	**End**
	Specification	1	41
	Claims	42	49
	Abstract	50	50

Warnings:					
Information:					
4	Drawings-only black and white line drawings	drawings.pdf	98804 a97d5bd4168fa5f5441deadd8ff4987a4c27c149	no	7
Warnings:					
Information:					
5	Oath or Declaration filed	Declaration198U2.pdf	101955 6cd38152f89d596aa40570765d3cd45821a5cf68	no	3
Warnings:					
Information:					
6		prelamend198U2.pdf	323820 33594cdb1ec1ecc8a639d775790d21a15f3684d7	yes	7

	Multipart Description/PDF files in .zip description		
	Document Description	**Start**	**End**
	Preliminary Amendment	1	1
	Claims	2	6

	Applicant Arguments/Remarks Made in an Amendment	7	7
Warnings:			
Information:			
	Total Files Size (in bytes):	2426879	

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

NON-PROVISIONAL
UTILITY PATENT APPLICATION
TRANSMITTAL - 37 CFR 1.53(b)

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Attorney Docket No.: I0113-198U2
First Named Inventor: Rocco Burgo
ELECTRONICALLY FILED
Applicant Name: Inolex Investment Corporation
Total Pages of Transmittal Form: 3

Transmitted herewith for filing is the non-provisional utility patent application entitled:

NATURAL 1,2-ALKANEDIOLS, COMPOSITIONS HAVING NATURAL 1,2-ALKANEDIOLS AND PROCESSES FOR MAKING THE SAME

which is:

an **[X]** Original; or

a [ ] Continuation, [ ] Divisional, or [ ] Continuation-in-part (CIP) of prior U.S. Non-Provisional Patent Application No. _____, filed ____.
Anticipated Group/Art Unit: ___ or Class _____, Subclass ____.

[X] This non-provisional patent application is based on Provisional Patent Application No. 62/623,985, filed January 30, 2018.

Enclosed are:

[X] Specification (including Abstract) and claims: 50 pages.

[X] 7 sheets of drawings (formal).

[X] Application Data Sheet Under 37 C.F.R. 1.76.

[X] Unexecuted: [ ] Inventor Declaration; [ ] Inventor Declaration and Power of Attorney; [ ] Inventor Oath; [ ] Substitute Statement.

[ ] Copy of [ ] Declaration; [ ] Declaration and Power of Attorney from prior application.

[ ] Copy of separate Power of Attorney (including 37 CFR 3.73(b) statement, if applicable).

[ ] Microfiche computer program (Appendix).

[ ] Nucleotide and/or Amino Acid Sequence Submission, including:

[ ] Computer readable copy [ ]Paper Copy [ ] Verified Statement.

[X] Name of Assignee: Inolex Investment Corporation

[ ] Certified copy(ies) of ____ Application No(s). _____ filed ____ is/are filed:

[ ] herewith or [ ] in prior application _____.

[ ] Applicant(s), by its/their undersigned attorney, claim(s) Small Entity Status under 37 C.F.R. §1.27 as [ ] an Independent Inventor, or [ ] a Small Business Concern, or [ ] a Non-Profit Organization.

[X] **Preliminary Amendment.**

[ ] Information Disclosure Statement, PTO/SB/08A or B, and cited references.

[ ] Request for Nonpublication of Application Under 35 U.S.C. §122(b)

[ ] Other:

The filing fee is calculated as follows:

			MICRO ENTITY			**SMALL ENTITY**			**LARGE ENTITY**	
CLAIMS	NO. FILED	NO. EXTRA	BASIC FILING, SEARCH, AND EXAMINATION FEES:	$430		BASIC FILING, SEARCH, AND EXAMINATION FEES:	$ 785		BASIC FILING, SEARCH, AND EXAMINATION FEES:	$1,720
Total	46-20 =	26	X $ 25	$	OR	X $ 50	$	OR	X $100	$2,600
Independent	2 - 3=	0	X $115	$	OR	X $230	$	OR	X $460	$920
[0] Multiple Dependent Claims			X $205	$	OR	X $410	$	OR	X $820	$0
[ ] Application Size Fee (if specification and drawings exceed 100 sheets)										
Total sheets	-100	Add'l sheets =	# of sets of 50 (round up to whole number) /50 =	X $100 =	$	OR	X $200 =	$	OR	X $400	$0
Late Fee For Filing Fees, Inventor Oath or Declaration			$ 40	$	OR	$ 80	$	OR	$160	$160
Late Fee For Non-English Specification			$ 35	$	OR	$ 70	$	OR	$140	$0
			TOTAL	$	OR	**TOTAL**	$	OR	**TOTAL**	$5400

[X] **The Commissioner is not authorized to charge the filing fee at this time as we elect to defer payment of the entire filing fee until receipt of a Notice to File Missing Parts**

[ ] Applicant herewith charges the above-noted filing fees in the amount of $___ as calculated above to the credit card account, Calderone Bullock American Express (Billing No. I0113.0251).

[X] The Commissioner is hereby authorized to charge and/or credit Deposit Account No. 60-2682 (Billing No. I0113.0251) as noted below.

[ ] Any overpayments or deficiencies in the above-calculated fee.

[ ] Any additional fees required under 37 C.F.R. § 1.16 and § 1.17.

[X] In the event that a Petition for Extension of Time is required during the prosecution of this application, but not submitted, please charge any extension fee under 37 C.F.R. § 1.136(a) to our Deposit Account noted above.

CORRESPONDENCE ADDRESS

1/30/19 By: _____
(Date)

LYNDA L. CALDERONE
Registration No. 35,837
CALDERONE BULLOCK LLC
Two Greentree Centre
9000 Lincoln Drive East, Suite 301
Marlton, NJ 08053
Telephone: 856-446-6047
Direct Dial: 856-446-6045
Facsimile: 856-267-5370
E-Mail: lcalderone@calderonebullock.com

[X] Customer Number or Bar Code Label: **000159443**

LLC:llc
Enclosures: Preliminary Amendment

PTO/AIA/14 (02-18)
Approved for use through 11/30/2020. OMB 0651-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

Application Data Sheet 37 CFR 1.76	Attorney Docket Number	I0113-198U2
	Application Number	
Title of Invention	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same	

The application data sheet is part of the provisional or nonprovisional application for which it is being submitted. The following form contains the bibliographic data arranged in a format specified by the United States Patent and Trademark Office as outlined in 37 CFR 1.76.
This document may be completed electronically and submitted to the Office in electronic format using the Electronic Filing System (EFS) or the document may be printed and included in a paper filed application.

Secrecy Order 37 CFR 5.2:

☐ Portions or all of the application associated with this Application Data Sheet may fall under a Secrecy Order pursuant to 37 CFR 5.2 (Paper filers only. Applications that fall under Secrecy Order may not be filed electronically.)

Inventor Information:

Inventor 1 Remove

Legal Name

Prefix	Given Name	Middle Name	Family Name	Suffix
	Rocco	V.	Burgo	

Residence Information (Select One) ● US Residency Non US Residency Active US Military Service

City	State/Province	Country of Residence
Mullica Hill	NJ	US

Mailing Address of Inventor:

Address 1	604 Widgeon Court
Address 2	

City	State/Province	Postal Code	Country
Mullica Hill	NJ	08062	US

Inventor 2 Remove

Legal Name

Prefix	Given Name	Middle Name	Family Name	Suffix
	Michael	E.	Wright	

Residence Information (Select One) ◉ US Residency Non US Residency Active US Military Service

City	State/Province	Country of Residence
Hurricane	UT	US

Mailing Address of Inventor:

Address 1	4175 S. 1200 W.
Address 2	

City	State/Province	Postal Code	Country
Hurricane	UT	84737	US

Inventor 3 Remove

Legal Name

Prefix	Given Name	Middle Name	Family Name	Suffix
	Gary	B.	Mosser	

Residence Information (Select One) ◉ US Residency Non US Residency Active US Military Service

Application Data Sheet 37 CFR 1.76	Attorney Docket Number	I0113-198U2
	Application Number	
Title of Invention	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same	

City	Tabernacle	State/Province	NJ	Country of Residence	US

Mailing Address of Inventor:

Address 1	6 Colonial Court		
Address 2			
City	Tabernacle	State/Province	NJ
Postal Code	08088	Country	US

Inventor 4 — Remove

Legal Name

Prefix	Given Name	Middle Name	Family Name	Suffix
	Michael	J.	Fevola	

Residence Information (Select One) ◉ US Residency ○ Non US Residency ○ Active US Military Service

City	Belle Mead	State/Province	NJ	Country of Residence	US

Mailing Address of Inventor:

Address 1	409 Township Line Road		
Address 2			
City	Belle Mead	State/Province	NJ
Postal Code	08502	Country	US

All Inventors Must Be Listed - Additional Inventor Information blocks may be generated within this form by selecting the **Add** button. Add

Correspondence Information:

Enter either Customer Number or complete the Correspondence Information section below.
For further information see 37 CFR 1.33(a).

☐ **An Address is being provided for the correspondence Information of this application.**

Customer Number	159443		
Email Address	USP@calderonebullock.com	Add Email	Remove Email

Application Information:

Title of the Invention	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same		
Attorney Docket Number	I0113-198U2	Small Entity Status Claimed	☐
Application Type	Nonprovisional		
Subject Matter	Utility		
Total Number of Drawing Sheets (if any)	7	Suggested Figure for Publication (if any)	1

PTO/AIA/14 (02-18)
Approved for use through 11/30/2020. OMB 0651-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

Application Data Sheet 37 CFR 1.76	Attorney Docket Number	I0113-198U2
	Application Number	
Title of Invention	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same	

Filing By Reference:

Only complete this section when filing an application by reference under 35 U.S.C. 111(c) and 37 CFR 1.57(a). Do not complete this section if application papers including a specification and any drawings are being filed. Any domestic benefit or foreign priority information must be provided in the appropriate section(s) below (i.e., "Domestic Benefit/National Stage Information" and "Foreign Priority Information").

For the purposes of a filing date under 37 CFR 1.53(b), the description and any drawings of the present application are replaced by this reference to the previously filed application, subject to conditions and requirements of 37 CFR 1.57(a).

Application number of the previously filed application	Filing date (YYYY-MM-DD)	Intellectual Property Authority or Country

Publication Information:

☐ Request Early Publication (Fee required at time of Request 37 CFR 1.219)

☐ **Request Not to Publish.** I hereby request that the attached application not be published under 35 U.S.C. 122(b) and certify that the invention disclosed in the attached application **has not and will not** be the subject of an application filed in another country, or under a multilateral international agreement, that requires publication at eighteen months after filing.

Representative Information:

Representative information should be provided for all practitioners having a power of attorney in the application. Providing this information in the Application Data Sheet does not constitute a power of attorney in the application (see 37 CFR 1.32).
Either enter Customer Number or complete the Representative Name section below. If both sections are completed the customer Number will be used for the Representative Information during processing.

Please Select One:	● Customer Number	US Patent Practitioner	○ Limited Recognition (37 CFR 11.9)
Customer Number	159443		

Domestic Benefit/National Stage Information:

This section allows for the applicant to either claim benefit under 35 U.S.C. 119(e), 120, 121, 365(c), or 386(c) or indicate National Stage entry from a PCT application. Providing benefit claim information in the Application Data Sheet constitutes the specific reference required by 35 U.S.C. 119(e) or 120, and 37 CFR 1.78.
When referring to the current application, please leave the "Application Number" field blank.

Prior Application Status	Pending		Remove
Application Number	Continuity Type	Prior Application Number	Filing or 371(c) Date (YYYY-MM-DD)
	Claims benefit of provisional	62623985	2018-01-30

Additional Domestic Benefit/National Stage Data may be generated within this form by selecting the **Add** button. Add

PTO/AIA/14 (02-18)
Approved for use through 11/30/2020. OMB 0651-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

Application Data Sheet 37 CFR 1.76	Attorney Docket Number	I0113-198U2
	Application Number	
Title of Invention	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same	

Foreign Priority Information:

This section allows for the applicant to claim priority to a foreign application. Providing this information in the application data sheet constitutes the claim for priority as required by 35 U.S.C. 119(b) and 37 CFR 1.55. When priority is claimed to a foreign application that is eligible for retrieval under the priority document exchange program (PDX)[i] the information will be used by the Office to automatically attempt retrieval pursuant to 37 CFR 1.55(i)(1) and (2). Under the PDX program, applicant bears the ultimate responsibility for ensuring that a copy of the foreign application is received by the Office from the participating foreign intellectual property office, or a certified copy of the foreign priority application is filed, within the time period specified in 37 CFR 1.55(g)(1).

Remove

Application Number	Country[i]	Filing Date (YYYY-MM-DD)	Access Code[i] (if applicable)

Additional Foreign Priority Data may be generated within this form by selecting the **Add** button. Add

Statement under 37 CFR 1.55 or 1.78 for AIA (First Inventor to File) Transition Applications

☐ This application (1) claims priority to or the benefit of an application filed before March 16, 2013 and (2) also contains, or contained at any time, a claim to a claimed invention that has an effective filing date on or after March 16, 2013.
NOTE: By providing this statement under 37 CFR 1.55 or 1.78, this application, with a filing date on or after March 16, 2013, will be examined under the first inventor to file provisions of the AIA.

Application Data Sheet 37 CFR 1.76	Attorney Docket Number	I0113-198U2
	Application Number	
Title of Invention	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same	

Authorization or Opt-Out of Authorization to Permit Access:

When this Application Data Sheet is properly signed and filed with the application, applicant has provided written authority to permit a participating foreign intellectual property (IP) office access to the instant application-as-filed (see paragraph A in subsection 1 below) and the European Patent Office (EPO) access to any search results from the instant application (see paragraph B in subsection 1 below).

Should applicant choose not to provide an authorization identified in subsection 1 below, applicant **must opt-out** of the authorization by checking the corresponding box A or B or both in subsection 2 below.

NOTE: This section of the Application Data Sheet is **ONLY** reviewed and processed with the **INITIAL** filing of an application. After the initial filing of an application, an Application Data Sheet cannot be used to provide or rescind authorization for access by a foreign IP office(s). Instead, Form PTO/SB/39 or PTO/SB/69 must be used as appropriate.

1. Authorization to Permit Access by a Foreign Intellectual Property Office(s)

A. Priority Document Exchange (PDX) - Unless box A in subsection 2 (opt-out of authorization) is checked, the undersigned hereby **grants the USPTO authority** to provide the European Patent Office (EPO), the Japan Patent Office (JPO), the Korean Intellectual Property Office (KIPO), the State Intellectual Property Office of the People's Republic of China (SIPO), the World Intellectual Property Organization (WIPO), and any other foreign intellectual property office participating with the USPTO in a bilateral or multilateral priority document exchange agreement in which a foreign application claiming priority to the instant patent application is filed, access to: (1) the instant patent application-as-filed and its related bibliographic data, (2) any foreign or domestic application to which priority or benefit is claimed by the instant application and its related bibliographic data, and (3) the date of filing of this Authorization. See 37 CFR 1.14(h)(1).

B. Search Results from U.S. Application to EPO - Unless box B in subsection 2 (opt-out of authorization) is checked, the undersigned hereby **grants the USPTO authority** to provide the EPO access to the bibliographic data and search results from the instant patent application when a European patent application claiming priority to the instant patent application is filed. See 37 CFR 1.14(h)(2).

The applicant is reminded that the EPO's Rule 141(1) EPC (European Patent Convention) requires applicants to submit a copy of search results from the instant application without delay in a European patent application that claims priority to the instant application.

2. Opt-Out of Authorizations to Permit Access by a Foreign Intellectual Property Office(s)

☐ A. Applicant **DOES NOT** authorize the USPTO to permit a participating foreign IP office access to the instant application-as-filed. If this box is checked, the USPTO will not be providing a participating foreign IP office with any documents and information identified in subsection 1A above.

☐ B. Applicant **DOES NOT** authorize the USPTO to transmit to the EPO any search results from the instant patent application. If this box is checked, the USPTO will not be providing the EPO with search results from the instant application.

NOTE: Once the application has published or is otherwise publicly available, the USPTO may provide access to the application in accordance with 37 CFR 1.14.

PTO/AIA/14 (02-18)
Approved for use through 11/30/2020. OMB 0651-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

Application Data Sheet 37 CFR 1.76	Attorney Docket Number	I0113-198U2
	Application Number	
Title of Invention	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same	

Applicant Information:

Providing assignment information in this section does not substitute for compliance with any requirement of part 3 of Title 37 of CFR to have an assignment recorded by the Office.

Applicant 1 — Remove

If the applicant is the inventor (or the remaining joint inventor or inventors under 37 CFR 1.45), this section should not be completed. The information to be provided in this section is the name and address of the legal representative who is the applicant under 37 CFR 1.43; or the name and address of the assignee, person to whom the inventor is under an obligation to assign the invention, or person who otherwise shows sufficient proprietary interest in the matter who is the applicant under 37 CFR 1.46. If the applicant is an applicant under 37 CFR 1.46 (assignee, person to whom the inventor is obligated to assign, or person who otherwise shows sufficient proprietary interest) together with one or more joint inventors, then the joint inventor or inventors who are also the applicant should be identified in this section.

Clear

Assignee	Legal Representative under 35 U.S.C. 117	Joint Inventor
Person to whom the inventor is obligated to assign.	Person who shows sufficient proprietary interest	

If applicant is the legal representative, indicate the authority to file the patent application, the inventor is:

Name of the Deceased or Legally Incapacitated Inventor:

If the Applicant is an Organization check here. ☒

Organization Name	Inolex Investment Corporation		
Mailing Address Information For Applicant:			
Address 1	103 Springer Building		
Address 2	3411 Silverside Road		
City	Wilmington	**State/Province**	DE
Country	US	Postal Code	19810
Phone Number		Fax Number	
Email Address			

Additional Applicant Data may be generated within this form by selecting the Add button. Add

Assignee Information including Non-Applicant Assignee Information:

Providing assignment information in this section does not substitute for compliance with any requirement of part 3 of Title 37 of CFR to have an assignment recorded by the Office.

PTO/AIA/14 (02-18)
Approved for use through 11/30/2020. OMB 0651-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

Application Data Sheet 37 CFR 1.76	Attorney Docket Number	I0113-198U2
	Application Number	
Title of Invention	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same	

Assignee 1

Complete this section if assignee information, including non-applicant assignee information, is desired to be included on the patent application publication. An assignee-applicant identified in the "Applicant Information" section will appear on the patent application publication as an applicant. For an assignee-applicant, complete this section only if identification as an assignee is also desired on the patent application publication.

Remove

If the Assignee or Non-Applicant Assignee is an Organization check here. ☐

Prefix	**Given Name**	Middle Name	**Family Name**	Suffix

Mailing Address Information For Assignee including Non-Applicant Assignee:

Address 1			
Address 2			
City		**State/Province**	
Country i		Postal Code	
Phone Number		Fax Number	
Email Address			

Additional Assignee or Non-Applicant Assignee Data may be generated within this form by selecting the Add button. Add

Signature:

Remove

NOTE: This Application Data Sheet must be signed in accordance with 37 CFR 1.33(b). **However, if this Application Data Sheet is submitted with the INITIAL filing of the application and either box A or B is not checked in subsection 2 of the "Authorization or Opt-Out of Authorization to Permit Access" section, then this form must also be signed in accordance with 37 CFR 1.14(c).**

This Application Data Sheet **must** be signed by a patent practitioner if one or more of the applicants is a **juristic entity** (e.g., corporation or association). If the applicant is two or more joint inventors, this form must be signed by a patent practitioner, **all** joint inventors who are the applicant, or one or more joint inventor-applicants who have been given power of attorney (e.g., see USPTO Form PTO/AIA/81) on behalf of **all** joint inventor-applicants.

See 37 CFR 1.4(d) for the manner of making signatures and certifications.

Signature	/Lynda L. Calderone/			Date (YYYY-MM-DD)	2018-01-30
First Name	Lynda L.	Last Name	Calderone	Registration Number	35837

Additional Signature may be generated within this form by selecting the Add button. Add

Application Data Sheet 37 CFR 1.76	Attorney Docket Number	I0113-198U2
	Application Number	
Title of Invention	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same	

This collection of information is required by 37 CFR 1.76. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 23 minutes to complete, including gathering, preparing, and submitting the completed application data sheet form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

Privacy Act Statement

The Privacy Act of 1974 (P.L. 93-579) requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1 The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C. 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether the Freedom of Information Act requires disclosure of these records.

2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.

3 A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.

4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).

5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent CooperationTreaty.

6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).

7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.

8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspections or an issued patent.

9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

TITLE OF THE INVENTION

[0001] Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making The Same.

CROSS-REFERENCE TO RELATED APPLICATIONS

[0002] This U.S. Non-Provisional Patent Application claims the benefit under 35 U.S.C. §119(e) of U.S. Provisional Patent Application No. 62/623,985, filed January 30, 2018, entitled, "Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Methods of Making Same," the entire disclosure of which is incorporated herein by reference.

BACKGROUND OF THE INVENTION

FIELD OF THE INVENTION

[0003] The invention herein relates to the field of natural and renewable, *i.e.*, sustainable, antimicrobial components that enable alternative preservation systems for use in compositions applicable to various industries, including personal care, household and institutional or industrial cleaning products, medical or other related uses, and particularly in personal care compositions. More particularly to the preparation of natural and renewable 1,2-alkanediols, methods for making natural and renewable 1,2-alkanediols and compositions including such 1,2-alkanediols prepared from natural and renewable ("sustainable") feedstocks.

DESCRIPTION OF RELATED ART

[0004] In most countries, including the United States, cosmetic and/or personal care ("personal care products") and other consumer products are manufactured and packaged in clean, but not typically sterile conditions, and many are sold in non-sterile forms. As a result, inconsequential (and usually harmless) amounts of bacteria, mold or yeast spores (collectively, "microbes") may be caught in products. In addition, once a consumer has purchased, opened and begun using such a product, further contamination may occur. For example, in personal care products, small amounts of microbes from the air or a consumer's skin may further contaminate the product.

[0005] Over time, during transit, storage or use the initial microbe population that was negligible may increase to a level significant enough to result in discoloration or fouling of the

product, including the appearance of visible mold, which can impact the usefulness or performance of the product. In the case of personal care products or other consumer products that are applied or ingested (such as vitamins, etc.), it can have a health impact as well, and in some circumstances can lead to adverse skin reactions upon application, including infection.

[0006] To address the issue of microbial growth, manufacturers in the personal care industry and in other industries commonly add chemical preservatives, which are typically petrochemically based, to such products and compositions. However, the type and amount of preservative that can be used is subject to restrictions on several fronts: (1) legislative (in some countries, use of particular preservatives are prohibited); (2) technical (the type/amount of preservative must demonstrate its efficacy by meeting certain empirically assessed criteria ("challenge testing"); (3) consumer preference (consumers may perceive certain preservative chemicals as undesirable, such as petroleum-based or non-sustainable products, so products including them are not commercial viable); (4) logistical (*e.g.*, the selected preservative must be effective in the relevant chemical environment (where parameters such as pH, hydrophobicity/hydrophilicity, etc. may vary) and production-cost friendly); and (5) sustainability, *i.e.*, it should be prepared using sustainable processes and feedstock sourcing as described further herein.

[0007] There is also a desire to develop alternatives to traditional preservation and to find more user-friendly chemical component combinations that are derived from sustainable and renewable resources (*i.e.*, "feedstocks"). These natural and renewable antimicrobial components and alternatives to traditional preservatives ("alternative preservation"), should provide equal or better protection and performance to the consumer.

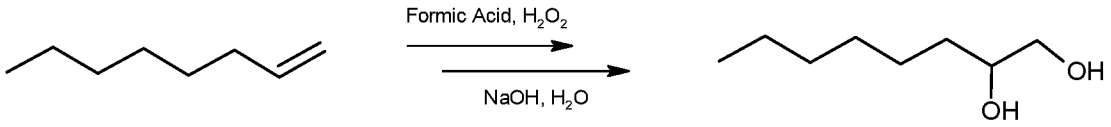
[0008] To resolve issues with traditional preservatives, the applicant herein developed a prior art preservative for personal care compositions that does not include parabens, formaldehyde donors or chlorinated compounds, that can be used in all types of personal care formulations (*e.g.*, both "leave-on" and "rinse-off" products). Such paraben-free or other non-petroleum-based alternative preservation products are attractive to consumers. The paraben-free preservative demonstrates efficacy against a broad spectrum of microorganisms at various levels of pH, especially around neutral pH levels which hare typically in or on the human body. This preservative is described in U.S. Patent Publication No. 2017/0360035 A1, in which one of the components in that preservative system is a 1,2-alkanediol.

[0009] Another attempt in the art to reduce the use of traditional preservatives is in the application of "hurdle technology." Hurdle technology is the practice of applying several types of restricting or preservative materials or processes so that microbes have a "hurdle(s)" to overcome before they can proliferate to spoilage levels (e.g., adding something to change pH to reduce proliferation and a smaller amount of preservative or other compound that together retard or create "hurdles" to microbial growth). Use of alternative preservation can be one step or "hurdle" in hurdle technology.

[0010] There is a need in the art to further develop "alternative preservation" additives. These alternative preservation systems include consumer-friendly, paraben-free materials that move away from traditional preservation or antimicrobials and towards "natural" and "sustainable" ("renewable") formulations and feedstocks, as those terms are defined hereinbelow.

[0011] One diol employed in preservation, caprylyl glycol, is believed by the applicant to be the fastest growing alternative preservation component in the world. Other 1,2-alkanediols such as 1,2-hexanediol, 1,2-decanediol, and 1,2-dodecanediol are also growing in popularity and function in alternative preservation formulations.

[0012] An example of one reaction sequence to prepare caprylyl glycol (1,2-octanediol) using an alkene as a key starting material is shown below. The starting material used is a petrochemically-derived 1-octene:



petroleum-based 1-octene

petroleum-based 1,2-octanediol

In the above reaction sequence, a petrochemically-derived 1-octene is reacted in the presence of formic acid and peroxide to form an intermediate chemical mixture which, e.g., undergoes a further ring-opening reaction in water with sodium hydroxide to complete formation of the 1,2-octanediol. This provides an excellent alternative preservation additive and is paraben-free, however, the starting material (*i.e.*, 1-octene) is derived from a fossil-based (petrochemical) resource and importantly, represents a non-renewable feedstock, *i.e.*, a non-sustainable resource.

[0013] Thus, as noted above there is a need for more consumer- and eco-friendly, alternative preservation systems (used alone or as part of hurdle technologies), including those that are paraben-free. There is further a need in the art for a material that is an alternative preservation material or that may be used in an alternative preservation system that is a 100% natural and renewable ("sustainable") feedstock to create an antimicrobial agent that can be used as or in alternative preservation systems. An example of a "natural" antimicrobial is one that is prepared from a bio-derived feedstock (*e.g.*, from current and sustainable agricultural activities, like non-GMO based-fermentation, algae-, plant- or vegetable-derived, *e.g.*, it comes from vegetable sources, or biomass, and it is not petrochemically-derived (such as being derived from sustainable tree and plant farms active in the 21st century). Such feedstocks are referred to herein as "natural" and "renewable" (*i.e.*, "sustainable") and are known as a non-petroleum-derived feedstock. Further, such materials are formed by "new" carbon and not from petroleum or other fossil fuel sources ("old" carbon). Such products are referred to herein as "natural" products and are known in the art as non-petrochemically-derived or "bio" products. By "sustainable," herein, the applicants are referring to materials that come from renewable sources, and not those that deplete a limited natural resource, such as a fossil fuel or other non-renewable resource such as petroleum. Thus a natural or bio product, which is not petrochemically derived, and/or is made from sources that are not petrochemically derived would be sustainable and renewable.

[0014] True natural products (bio-compounds) are formed using biomass (*e.g.*, material stored from carbon cycle processes in living plants and roots, etc. or released through animal respiration or through decomposition). When carbon decomposes and is broken down over millions of years under pressure, it creates fossil fuels (the source of petrochemically-derived carbon). Bio-compounds herein are intended to include materials derived from the carbon of plant sources/biomass, that exist recently and are also sustainable, and are not derived from fossil fuels.

[0015] These bio-based or "natural" feedstocks may be used in the production of alternative preservation formulations. The bio-based or "natural" products from such feedstocks may be tested to determine that they come from a true, natural and sustainable (as those terms are defined herein) feedstock source. Some products are known or advertised as being from natural sources when, in fact, they may not be prepared from truly natural and/or sustainable feedstock. A natural organic product is typically defined as a compound produced naturally by a living

organism. To distinguish a petroleum-based product from a truly natural and/or sustainable product, one must test for the authenticity using established and credible test methods. The most current method employs a detailed analysis of stable isotopes using mass spectroscopy and evaluating carbon-12/carbon-13 and/or hydrogen-1/hydrogen-2 ratios. Such testing is available through several analytical service testing organizations and is much faster, more cost effective, and yields more detailed information compared to radiocarbon testing methods.

[0016] Stable isotope analysis is based on the principle of kinetic isotope effect. The latter effect is well-known to those in the art of chemical kinetics. In the broadest terms, heavy isotopes of a particular element react slower than their lighter equivalent (*e.g.*, carbon-12 as opposed to carbon-13). So, as plants incorporate carbon dioxide into their biomass, the ratio of carbon-12 to carbon-13 will vary depending on the type of chemistry used in the plant to make biomass (*e.g.*, whether the plant undergoes a C_3 or C_4 photosynthesis pathway). This is commonly reported as the $\delta^{13}C/^{12}C$ ratio (*i.e.*, $\delta^{13}C$), and is referenced to a current carbon dioxide standard. In addition, similar isotope kinetic effects are observed when water is incorporated into new biomass, and this is measured as the $\delta^{2}H/^{1}H$ ratio (*i.e.*, $\delta^{2}H$). Using a combination of $\delta^{13}C$ and $\delta^{2}H$ ratios, one familiar with in the relevant art is able to readily distinguish and validate the nature of the feedstock that was used to prepare the product being analyzed (*i.e.*, whether it is petrochemically-derived or derived from recently living or living algae-, plant- or similar bio-sources).

[0017] In Fig. 2, it can be seen in general terms how isotope ratios can be used to determine the source of various detergents that have a strong relation to the feedstocks described herein, like 1-octanol. From the plot in Fig. 2, the $\delta^{2}H$ values have a more clearly defined difference between petroleum-based and renewable feedstocks and so in this case prove more valuable and defining than the $\delta^{13}C$ values. However, the combination using $\delta^{2}H$ and $\delta^{13}C$ values together is a preferred technique employed in the invention herein to prove a feedstock is, in fact, natural and renewable.

[0018] Radiocarbon is an unstable isotope of carbon, known as ^{14}C. ^{14}C is an unstable isotope that emits radiation energy in the form of *beta* particles at a very consistent rate and

ultimately decays to the more stable ^{14}N (*i.e.*, a half-life for radiocarbon is 5730 years). Because petroleum-based (*i.e.*, petrochemically-derived) feedstocks are derived from plants and animals buried millions of years ago, the feedstocks' radiocarbon (*i.e.*, ^{14}C) has been lost to decay. The ASTM International standards provide testing standards to determine the authenticity of a "bio-based compound" using radiocarbon, which may be found in ASTM D6866-16. This standard distinguishes newer carbon from carbon derived from fossil-fuel, or petroleum- and petrochemically-derived sources, *i.e.*, "old carbon." The amount of ^{14}C in recent or current biomass is known, so a percentage of carbon from a renewable source can be estimated from a total organic carbon analysis, which provides the data necessary to determine if a compound is truly derived from a "natural" and/or "sustainable" ("renewable") feedstock source or is derived conversely from a compound of "old" sequestration (*i.e.*, a petrochemically-derived or petroleum-based source). The use of petroleum-based or often labeled fossil-based feedstocks is generally accepted as being non-sustainable, *i.e.*, old carbon is a non-sustainable and not a renewable feedstock, and furthermore, is not considered "natural" and "sustainable" in the art. As defined herein, it would not be considered to be a "natural" product or useful in a "natural" formulation. Hence, use of such feedstocks does not represent a path forward to development of "natural" and "renewable" alternative preservatives.

[0019] Dehydration of terminal alcohols using traditional methods to form alkene starting materials chemically yields a mixture of 1- alkenes, 2- alkenes, and 3- alkenes due to rearrangements and migrations of the 1-alkene product during the dehydration process. Using such a mixture of alkenes ultimately provides a mixture of diols upon dihydroxylation, providing a low yield of the desired terminal 1,2-diol material and unwanted contaminants.

[0020] In one known path of forming caprylyl glycol (1,2-octanediol) starting with 1-octanol, made from petrochemically-derived (*i.e.*, petroleum-derived or fossil fuel-derived) feedstocks, the following reaction is known to occur:

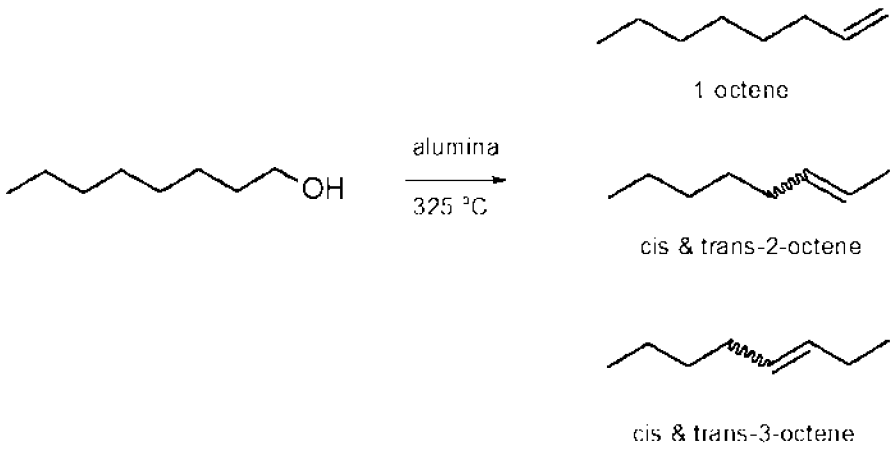
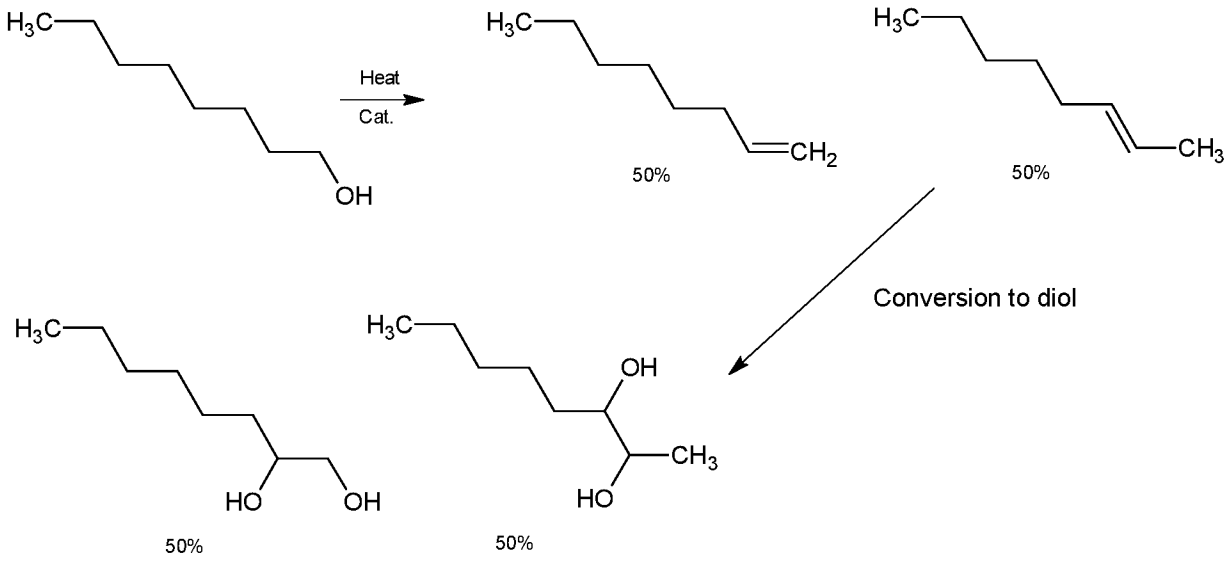


Table A

Catalyst	1-Octene (%)	2-Octene & 3-Octenes (%)	1-Octanol (%)	1-Octene Selectivity (%)
Alumina	87.7	5.3	4.7	94.3



As can be seen from the above-reaction schemes, the use of heat and a traditional acid catalyst converts 1-octanol to an alkene, *i.e.*, octene. Exposure to the acid sites in the catalyst allows the more thermodynamically stable 2-octene to form along with 1-octene. The 2-octene regioisomers formed (including 3-octene as shown above and 4-octene (not shown)) are very

difficult to separate from the 1-octene and afford a lower net chemical yield of the desired 1-octene. It follows that an increased or higher regioselectivity is a critical factor in the catalytic dehydration of 1-octanol to 1-octene and a desired goal to achieve in the relevant art. When using renewable and natural feedstocks, reaction efficiency (*i.e.*, chemical yield) of the desired product (*i.e.*, 1-alkenes) is critical. There remains a need in the art for a highly regioselective dehydration of 1-octanol that also has high chemical conversion using efficient process equipment and operating in a rapid and energy efficient manner.

[0021] Thus, traditional pathways using an octene mixture of regioisomers as shown above for conversion to a desired terminal 1,2-octanediol is reduced and the yield is also reduced. This makes the ability to form the desired alternative preservative materials chemically inefficient and not cost effective, creates more waste in process, and makes the overall process less sustainable. There is a strong need in the art for a highly regioselective, and high-chemical conversion rate process for creating a bio-1-octene from a bio-1-octanol.

[0022] International patent Publication No. WO 2004/078336 A2 uses a γ-alumina having no additional promoters but that was instead given a very large pore size and volume to enhance regioselectivity for dehydration of 1-octanol. In this work, the regioselectivity was high (97.7%), but, the chemical conversion was 65% and product selectivity for octenes was only 25%. Thus, the chemical yield of 1-octene from 1-octanol using this catalyst system was only about 15.9%.

[0023] In another example, U.S. Patent No. 7,576,250 B2, formation of 1-octene by cracking an ether derivative of 1-octanol is described. In this patent, the 1-octanol must first be converted to the methyl ether (*i.e.* methyl octyl ether) and then the ether is subjected to and passed over a γ-alumina catalyst. This involves extra chemical and added cost to make the ether and loss of the methanol in the process leading to a very ineffective and atom inefficient method. Most notably, the catalyst goes from having good conversion and regioselectivity in the first few hours, then, in just a mere 20 h of time-on-stream ("TOS"), all catalysts disclosed in the art show a constant and significant decrease in chemical conversion with a concomitant loss in regioselectivity. This type of short catalyst life time is not acceptable in commercial production efforts.

[0024] A lead-containing alumina catalyst is described in Chinese Patent No. 105312044 B, wherein plumbite pseudoboehmite showed high apparent selectivity and conversion. The catalyst had a carbon deposition rate of ~0.04%/h, which is highly undesirable and would be

unsatisfactory for something like 1000 h of continuous operation. Even more limiting is these catalysts incorporate a toxic heavy metal, namely lead. The use of these catalysts would require costly environmental controls and worker safety precautions if used on the industrial scale for commercial production

[0025] One path to making a "natural" alkanediol is by fermentation of biomass to form a straight chain, linear alkanediol. For example, U.S. Patent Application Publication No. 2005/0069997 A1 teaches formation of 1,3 propanediol from fermentation broth. However, 1,3-propanediol is not a 1,2-alkanediol. Fermentation processes that form long chain diols are chemically inefficient (*i.e.*, poor life-cycle-analysis numbers) and additionally require use of GMO-bacteria or -yeast, especially if one attempts to make a long carbon 1,2-alkandiol.

[0026] Another example of a natural bio-alcohol that is described in the art is bio-1-butanol, a known substance that has been used in commercial production since the discovery by Pasteur in 1862 of the acetone-butanol-ethanol fermentation process carried out by *clostridum* bacteria (a non-GMO bacteria), although it is used in very limited quantities currently. These bacteria ferment both C_5 and C_6 sugars to form a mixture of acetone, 1-butanol and ethanol (*i.e.*, ABE). Since the discovery of ABE, advances in fermentation processes can provide optimized production of bio-1-butanol over acetone and ethanol production. While the production is improved, it remains difficult and energy intensive to purify the bio-1-butanol from the fermentation broth and other impurities involved in the fermentation process such as ethanol and acetone. In addition, applicant is unaware of a non-GMO bacteria or yeast process that can form longer chain alcohols (*i.e.*, chains of six or more such as 1-hexanol or 1-octanol).

[0027] Bio-1-butanol is known as a source to make biofuels and provides a pathway for converting straight-chain, linear, primary four-carbon alcohols to bio-1-alkenes at very high yield to improve biofuel production and can also be used, for example in production of eco-friendly tires. In this method, bio-1-alcohols are dehydrated to bio-1-alkenes with high selectivity and chemical yield. The resulting bio-1-alkenes are useful in preparing high flashpoint diesel and jet biofuels useful for civilian and military applications. The bio-1-butanol is dehydrated using a solid phase dehydration catalyst that includes an inorganic support, such as γ-alumina or zinc aluminate ($ZnAl_2O_4$), and that has been treated/modified with a basic aqueous solution. The support may be further treated with an organosilane diluted in at least one hydrocarbon solvent. The process can convert bio-1-butanol from a fermentation

process that contains 0.1 to about 90 wt% water and produces a corresponding 1-alkene at a 92-99% regioselectivity, with only a single pass over the solid-phase catalyst, produing a reaction conversion of greater than 95% and a chemical yield of greater than 90% of the bio-1-butene. However, even with such a result for bio-1-butene, it is noted that the conversion of 1-octanol to 1-octene is not simply a homolog of a conversion of 1-butanol to bio-1-butene. In fact, comparative examples described herein below, illustrated that silanization of the catalyst leads to a *loss* of regioselectivity for the dehydration chemistry of 1-octanol.

[0028] Based on the foregoing, while progress has been made, there is a need in the art for continued improvement to consumer products such as personal care products, household products, industrial products and pharmaceutical products to provide additives, especially for antimicrobial use or for boosting the effects of known preservatives or alternative preservation systems, so as to keep products safe from contamination and fresh in storage and in use, while avoiding traditional preservatives and finding and relying on components for consumer product compositions from natural bio-sources. There is more particularly a need for a natural, and prefereably renewable/sustainable feedstock source to create a natural, bio-caprylyl glycol (*i.e.*, a natural or bio-1,2-octanediol) and the associated bio-1-alkene intermediate.

BRIEF SUMMARY OF THE INVENTION

[0029] The invention provides efficient pathways to achieve a 100% "natural" or "bio-compound" that is an authentic "natural" material, and preferably includes bio-1,2-alkanediols of carbon lengths from about 5 to about 20, and preferably from 5 to about 14, that can be used alone, in combinations of different types of bio-1,2-alkanediols, or in blends with other natural or traditional antimicrobials and other cosmetic additives.

[0030] The invention includes a process for the synthesis of bio-1,2-alkanediols, comprising: providing a bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%; and converting the bio-alkene to a bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms.

[0031] In one embodiment of the process, the bio-alkene and the bio-1,2-alkanediol each have a carbon chain of about 6 to about 14 carbon atoms. The bio-alkene and the bio-1,2-alkanediol each may have a carbon chain length of about 6 to about 10 carbon atoms. The bio-alkene and the bio-1,2-alkanediol each may have a carbon chain length of about 6 to about 8

carbon atoms. For example, the bio-alkene may be bio-octene and the bio-1,2-alkane diol may be bio-1,2-octanediol.

[0032] In a further embodiment of the process, the bio-alkene preferably results from a process wherein a bio-1-alcohol is dehydrated by heating the bio-1-alcohol in a reactor with a catalyst. The reactor may be a fixed bed reactor. The fixed bed reactor may also be, *e.g.*, a fluidized fixed bed reactor. In such an embodiment, the catalyst may be selected from $ZnAl_2O_4$ and a γ-alumina catalyst.

[0033] The bio-alkene in a preferred embodiment has a bio-1-alkene regioselectivity of about 92% to about 99%, and preferably about 95% to about 99%. The process also preferably yields at least about 92% to about 99% of the bio-1,2-alkanediol.

[0034] As noted above in one embodiment a catalyst treatment may be used. The catalyst may be a γ-alumina catalyst treated with a base to form a modified γ-alumina catalyst. The base may comprise a Group I or a Group II metal. The γ-alumina catalyst may be treated, *e.g.*, with a calcium promoter to provide a modified γ-alumina catalyst. The modified γ-alumina catalyst may then be calcined to provide a calcined γ-alumina catalyst. In one embodiment, wherein a calcium promoter is used, the calcium promoter may be used in an amount of about 0.01 weight percent to about 4 weight percent based on the weight of CaO determined after calcination, and preferably about 1 weight percent to about 2 weight percent based on the weight of CaO determined after calcination.

[0035] A preferred temperature during calcination is about 400°C to about 500°C, more preferably about 420°C to about 480°C, and most preferably about 440°C to about 460°C. Calcination may be carried out in an oven. The calcination may be performed in air or under an atmosphere of nitrogen or other inert gas(es).

[0036] The bio-alkene preferably undergoes a distillation step to refine the bio-alkene prior to converting the bio-alkene to the bio-1,2-alkanediol. The bio-1,2-alkanediol formed from the bio-alkene may also undergo a final distillation step to refine the bio-1,2-alkanediol.

[0037] In another embodiment of the process, the bio-alkene may result from a process wherein the bio-1-alcohol in water is dehydrated by heating the bio-1-alcohol with a catalyst using no purge gas or under a purge gas. The purge gas may preferably be nitrogen but other inert gases may be used within the scope of the invention.

[0038] The bio-alkene in the process may also be converted to the bio-1,2-alkanediol by reacting the bio-alkene in the presence of at least one of peracid, *e.g.*, formic acid or acetic acid,

and hydrogen peroxide to form a mixture of bio-1,2-epoxyalkane having an epoxy ring and bio-1,2-alkanediol, and contacting this mixture with water and sodium hydroxide to complete formation to the bio-1,2-alkanediol.

[0039] The overall process preferably process yields at least about 60% to about 99% of the bio-1,2-alkanediol, and more preferably yields at least about 72% to about 99% of the bio-1,2-alkanediol.

[0040] The invention further includes a process for making bio-1,2-alkanediols, comprising: providing a bio-1-alcohol and a catalyst treated with a base; dehydrating the bio-1-alcohol in the presence of the catalyst to form a bioalkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%; and converting the bio-alkene to a bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms. Preferably, the catalyst is treated with a promoter and then calcined.

[0041] The catalyst in this process may also be a γ-alumina catalyst and the base may comprise calcium. The γ-alumina catalyst is preferably treated with a calcium promotor and after treatment is calcined at a temperature of about 400°C to about 500°C. The process preferably yields about 92% to about 99% of the bio-1,2-alkanediol.

[0042] The invention further includes a method for treating a catalyst for use in dehydration of alcohols, comprising providing a γ-alumina catalyst; treating the γ-alumina catalyst with a base, preferably a promoter as described herein, comprising a Group I or a Group II metal; and heating the γ-alumina catalyst to a temperature of about 400°C to about 500°C, which is preferably done in a very controlled and deliberate manner. The base may comprise calcium, and preferably the base may be a calcium promoter and the heating occur during calcination. The calcium promoter may be used in an amount of about 0.01 weight percent to about 4 weight percent based on the weight of CaO determined after calcination, and more preferably in an amount of about 1 weight percent to about 2 weight percent based on the weight of CaO determined after calcination, preferably after careful calcination. The temperature during calcination in a preferred embodiment may be about 420°C to about 480°C, and more preferably may be about 440°C to about 460°C. Beneficial results are achieved based on the applicant's process including the preferred ranges noted. Applicant's determined that high calcination temperatures decrease catalyst regioselectivity and thus chemical yield of bio-1-alkene.

[0043] The invention further includes a composition, comprising at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

[0044] The composition may be a personal care composition, such as a hair care composition, an oral care composition, a skin care composition or a cosmetic composition.

[0045] The composition may also be a composition for a household product, such as a fabric care product or a cleaning product.

[0046] The composition may also be an industrial composition, or a pharmaceutical composition, a vitamin composition, or a health care composition.

[0047] In one embodiment of the composition herein, the composition may comprise a second bio-1,2-alkanediol, different than the first bio-1,2-alkanediol, the second bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a second bio-alkene, preferably regioselectively, such that the resulting bio-alkene of about 5 to about 20 carbon atoms has a terminal bio-alkene content of at least about 80% and a bio-1-alkene regioselectivity of at least about 80%. The composition may comprise at least one other bio-compound different from the first and the second bio-1,2-alkanediols. In another embodiment, the composition may comprise an antimicrobial compound different from the first and the second bio-1,2-alkanediol and different from the other bio-compound. The composition may also further comprise at least one other bio-compound different from the at least one bio-1,2-alkanediol, and in another embodiment, such a composition may also further comprise at least one antimicrobial compound different from the at least one bio-1,2-alkanediol and different from the other bio-compound. The first bio-1-alkene in such composition preferably has a regioselectivity of about 92% to about 99% and more preferably about 95% to about 99%. The first bio-alkene may be a bio-octene and the at least one first bio-1,2-alkane diol may be a bio-1,2-octanediol.

[0048] Also included in the invention is a method for providing an antimicrobial effect to a composition, including incorporating an antimicrobial system to the composition, wherein the antimicrobial system comprises at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of at least one first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%. The antimicrobial system may further comprise a

second bio-1,2-alkanediol different than the first bio-1,2-alkanediol, wherein the first bio-1,2-alkanediol has a carbon chain length of about 5 to about 20 carbon atoms and is formed by regioselectively converting a second bio-alkene, different than the first bio-alkene, having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%. The composition may further comprise at least one other bio-compound different from the first and the second bio-1,2-alkanediols, and in another embodiment may comprise at least one other preservative compound different from the first and the second bio-1,2-alkanediol and different from the other bio-compound. Such a composition may also further comprise at least one other bio-compound different from the at least one first bio-1,2-alkanediol. The antimicrobial may also further comprise at least one other antimicrobial compound and/or preservative compound different from the at least one first bio-1,2-alkanediol and different from the other bio-compound. The antimicrobial system may be made so as to demonstrate antimicrobial efficacy. The first bio-1-alkene preferably has a regioselectivity of about 92% to about 99%, and preferably about 95% to about 99%. The first bio-alkene may be bio-octene and the at least one first 1,2-alkane diol may be a bio-1,2-octanediol.

[0049] The invention further includes in one embodiment a method for boosting the antimicrobial efficiency of an antimicrobial and/or a preservative in a composition, comprising incorporating an antimicrobial system into the composition, wherein the second antimicrobial system comprises at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by regioselective conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%. In the method, the antimicrobial system may further comprise a second bio-1,2-alkanediol different than the first bio-1,2-alkanediol, wherein the second bio-1,2-alkanediol has a carbon chain length of about 5 to about 20 carbon atoms and is formed by conversion of a second bio-alkene, different than the first bio-alkene, having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%. In the composition, it may further comprise at least one other bio-compound different from the antimicrobial and/or preservative and different from the first and the second bio-1,2-alkanediols. The composition may also further comprise at least one other bio-compound different from the at least one first bio-1,2-alkanediol. The first bio-1-alkene preferably has a regioselectivity of about 92% to about 99%., and more preferably about 95% to about 99%. In one embodiment, the antimicrobial system demonstrates antimicrobial efficacy. The first bio-

alkene may be a bio-octene and the at least one first bio-1,2-alkane diol may be a bio-1,2-octanediol.

[0050] In yet a further embodiment, the invention includes an antimicrobial product, comprising at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%. The product may further comprise a second bio-1,2-alkanediol, different than the at least one first bio-1,2-alkanediol, the second bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms and formed by conversion of a second bio-alkene, different than the first bio-alkene, having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

[0051] The antimicrobial product may further comprise at least one other bio-compound different from the at least one first and the second bio-1,2-alkanediols. In one embodiment, the product may further comprise a second antimicrobial and/or a preservative different from the antimicrobial product. The product may also further comprise at least one other bio-compound different from the at least one first bio-1,2-alkanediol, and may also further comprise a second antimicrobial and/or a preservative different from the antimicrobial product. The first bio-1-alkene preferably has a regioselectivity of about 92% to about 99%.

[0052] The antimicrobial product preferably also demonstrates antimicrobial efficacy. The first bio-alkene may be bio-octene and the at least one first 1,2-alkane diol may be a bio-1,2-octanediol.

[0053] The invention also includes a product for boosting the efficacy of an antimicrobial and/or a preservative in a composition, the product comprising at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%. Such product may further comprise a second bio-1,2-alkanediol, different than the at least one first bio-1,2-alkanediol, the second bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms and formed by conversion of a second bio-alkene, different than the first bio-alkene, having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%. The composition of the product may also further comprise at least one other bio-compound different from the at least one first and the second

bio-1,2-alkanediols, and also may further comprise at least one other bio-compound different from the at least one first bio-1,2-alkanediol. The first bio-1-alkene preferably has a regioselectivity of about 92% to about 99%. The first bio-alkene may be bio-octene and the at least one first bio-1,2-alkane diol may be bio-1,2-octanediol. The product preferably demonstrates antimicrobial efficacy.

BRIEF DESCRIPTION OF THE SEVERAL VIEWS OF THE DRAWINGS

[0054] The foregoing summary, as well as the following detailed description of preferred embodiments of the invention, will be better understood when read in conjunction with the appended drawings. For the purpose of illustrating the invention, there is shown in the drawings embodiments which are presently preferred. It should be understood, however, that the invention is not limited to the precise arrangements and instrumentalities shown. In the drawings:

[0055] Fig. 1 is a process flow chart of steps in a preferred embodiment of a process described herein;

[0056] Fig. 2 is graphical representation of $\delta^{2}H$ and $\delta^{13}C$ values for petroleum-based alcohols, alkenes and acids and the corresponding natural and renewable analogs;

[0057] Fig. 3 is the Gas Chromatograph (GC) chromatogram of the reactor output from the dehydration of bio-1-octanol using the catalyst prepared in Example 4 at 0.2ml of bio-1-octanol feed and heated to 315°C;

[0058] Fig. 4 is the GC chromatogram of the bio-1-octene obtained from the reactor after distillation;

[0059] Fig. 5 is the GC chromatogram of the bio-1,2-octanediol obtained from dehydration of bio-1-octene, conversion to the 1,2-octanediol and then fractional distillation of the 1,2-octanediol;

[0060] Fig. 6 is the GC chromatogram of the petro-1,2-octanediol currently approved for use in cosmetic formulation; and

[0061] Fig. 7 is a photo comparing catalyst with 1400 h TOS (left) and new catalyst (on right).

DETAILED DESCRIPTION OF THE INVENTION

[0062] The invention includes a process for the synthesis of bio-1,2-alkanediols. As used herein, "bio" when used with "alkanediol," or any specific alkanediol compounds noted within this class of compounds is intended to mean as described above that the compound, and preferably all of the carbons in the compound within this class of compounds, as described further below are "natural" compounds that are also authentic natural compounds derived from plant sources (algae-, plant-, animal- or biomass-derived) and are non-petrochemically derived (*i.e.*, not derived from a non-renewable resource such as fossil fuel, petroleum or other "old" carbon source). In this sense, as they are derived from renewable materials, they are also sustainable compounds. It also means that such materials are formed from bio-alkenes. When used in the terms, "bio-alcohol," or "bio-alkene," and similar compounds herein, "bio" is intended also to have the same connotation as noted above for describing "bio-1,2-alkanediol." Such materials should preferably satisfy the requirements of having and being derived from an authentic or "new" carbon material (non-petrochemically derived) feedstock as confirmable through testing such a compound using mass spectroscopy as described above and/or gas chromatography, the current ASTM D6866-16 standard or prior methods such as specific stable isotope analysis (CSIA). The invention further includes the bio-1-alkenes and bio-1,2-alkanediols resulting from the synthesis process, as well as various products incorporating them. The invention further includes a method for treating a catalyst, which may be used in embodiments herein for dehydrating a bio-1-alcohol to form a bio-alkenes used in the processes herein for forming bio-1,2-alkanediols.

[0063] The invention provides efficient pathways to 100% "natural" or "bio" compounds that are from new carbon or authentic carbon materials that are not petrochemically derived and are preferably used in compositions and products that can be made for a wide variety of end uses and compositions, including as components of antimicrobial compositions and as cosmetic additives, among other uses described herein.

[0064] Also disclosed herein are methods of using bio-1,2-alkanediols for use as antimicrobials in a composition such as in or with an alternative preservation system and/or as hurdle technology. Also disclosed are various compositions comprising bio-1,2-alkanediols, including when used as an antimicrobial Also described are methods of boosting the antimicrobial efficacy of a composition already including a preservative or alternative

preservation system, and the resulting boosted antimicrobial efficacy composition. Each of such methods includes incorporating into the composition a bio-1,2-alkanediol, combination(s) of at least two different bio-1,2-alkanediols, or combination(s) of at least one bio-1,2-alkanediol with at least one other bio-compound different from the at least one bio-1,2-alkanediol, wherein the bio-1,2-alkanediol(s) either act as a antimicrobial or boost the efficacy of another antimicrobial, preservative or other alternative preservation system already present in a composition. The bio-1,2-alkanediols may themselves be an antimicrobial product and/or a antimicrobial efficacy boosting product whether used alone, in combinations of one or more different bio-1,2-alkanediols, or in combination(s) of one or more different bio-1,2-alkanediols with at least one other bio-compound different from the bio-1,2-alkanediols and/or an optional, different antimicrobial, preservative or alternative preservation system. The bio-1,2-alkanediols when used in compositions herein exhibit antimicrobial efficacy.

[0065] In one embodiment herein, a process is described herein for synthesis of bio-1,2-alkanediols. As noted above, materials referred to herein as "natural" are intended to include materials that are non-petrochemically derived and are sustainable, whether synthesized from non-petrochemically derived, natural materials or formed from a natural, and preferably an authentic natural or bio-based source.

[0066] The method includes providing a bio-alkene having a carbon chain of about 5 to about 20 carbon atoms, preferably about 6 to about 14 carbon atoms, and more preferably about 6 to about 10 carbon atoms, with a bio-1-octene being preferred. The bio-alkene is preferably a bio-1-alkene with a traditional α-olefinic structure. It also preferably has a regioselectivity when prepared of at least about 80%, more preferably at least about 92% and most preferably at least about 95% up to as much as 99%.

[0067] Such bio-alkenes may be bio-1-pentene, bio-1-iso-pentene, bio-1-hexene, bio-1-heptene, bio-1-octene, bio-1-nonene, bio-1-decene, bio-1-dodecene, and related bio-1-alkenes up to chain length of 20 carbons. Larger or smaller bio-alkenes may also be made, however, the preferred bio-alkenes for use herein are those with sufficient chain length to serve as effective antimicrobials when converted to bio-1,2-alkanediols without losing the desired properties. Such bio-alkenes may be straight or branched chain molecules, although straight chain molecules are preferred and may include one or more functional or substituted groups for desired end effects in the end use application, for example, sulfonate groups, hydroxyl groups, ether groups, amide groups, carbonyl groups, carboxylic acid groups, amine groups, fluorinated

groups, aryl or arene groups, and similar functional groups without intending to be limiting. Preferably any such functional or substituted groups do not interfere with conversion to an alkanediol or with the antimicrobial end application desired for the particular alkanediol.

[0068] Such bio-alkenes, such as bio-1-hexene and bio-1-octene, as are generated according to the invention, may be used to make bio-1,2-alkanediols as described herein, and may also be used to make other bio-based compounds and polymers, including, *e.g.*, for synthesis of bio-based olefin copolymers. Bio-1-alkenes herein may be used, for example, in free radical or catalytic polymerization processes as are known in the art, to yield a variety of bio-polyalphaolefins homopolymers and copolymers, which are useful as lubricants and cosmetic ingredients. A further example, includes bio-linear low-density polyethylene (bio-LLDPE) can be prepared via copolymerization of bio-ethylene (derived via dehydration of sugarcane derived bio-ethanol to bio-ethylene) with bio-1-alkene copolymers yielding 100% renewable bio-LLDPE, a thermoplastic resin useful in film applications such as food and non-food packaging and shrink/stretch films.

[0069] The process of the invention includes an embodiment wherein the process makes bio-1,2-alkanediols, including providing a bio-1-alcohol and a catalyst treated with a base, preferably with a promoter and then calcined; dehydrating the bio-1-alcohol in the presence of the catalyst to form a bioalkene having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%; and converting the bio-alkene to a bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms.

[0070] An overall flow diagram of a preferred process of making bio-1,2-alkanediols, that incorporates a preferred method of treating a catalyst as described further below is provided as embodiment 100 as shown in Fig. 1.

[0071] In the process generally referred to herein as process 100, in Fig. 1, prior to introducing the bio-1-alcohol, a catalyst is prepared. A suitable γ-alumina catalyst, e.g., Al_2O_3, 110 is introduced into a reactor 101 into which a calcium promotor or other base treatment material 102 as described further below is introduced. After a suitable treatment described in detail below, a modified catalyst 103 leaves reactor 101 and enters a heated chamber 104 such as an oven for heat treatment. If a catalyst promoter is used, for example, a calcination step occurs at preferred and controlled temperature ranges to increate a level of regioselectivity in a resulting bio-alkene in the next step. After, e.g., calcination in chamber 104, the calcined or otherwise heat-treated catalyst 105 is introduced along with a bio-1-alcohol 106 such as bio-1-

hexanol or bio-1-octanol into a reactor 107 to promote intimate contact between the alcohol and the catalyst. Suitable reactors included fixed bed reactors, fluidized bed reactors, tubular reactors, and other suitable reactors. After sufficient time for dehydration of the bio-1-alcohol to a bio-alkene 109, which highly regioselects to a bio-1-alkene, a further, optional distillation or other purification or refining step in a column(s) or other equipment 110, may occur. The refined or otherwise purified and highly regioselective bio-alkene, which should be of a high quantity of bio-1-alkene, is then introduced to a reactor 112 in order to convert the bio-1-alkene into a bio-1,2-alkane diol 113. Such bio-1.2-alkane diol 113 may be further and optionally refined such as by distillation in a column(s) or through other steps in equipment 114. After purification or distillation, a final high yield of up to 99% is removed as the final bio-1,2-alkane diol 115.

[0072] The bio-1-alkene may be made using a catalytic process, and preferably using a catalyst that has been treated according to the method described herein. In one embodiment, the bio-alkene, which is preferably made herein to be of a high regioselectivity bio-1-alkene is formed using a bio-1-alcohol that is dehydrated by heating the bio-1-alcohol in the presence of a catalyst. This may be done, for example, by passing the bio-1-alcohol through a fixed bed reactor, or a fluidized bed reactor. The fixed bed may be packed with a catalyst, preferable a treated catalyst having been treated using techniques as described below.

[0073] For example, a bio-1-alcohol derived from a plant source, including biomass that can produce a related acid (*e.g.*, 1-octanol may be derived from coconut, palm or other plant source or biomass that can produce octanoic acid, which is subsequently reduced via catalytic dehydrogenation to 1-octanol). The bio-1-alcohol is then dehydrated regioselectively by feeding the material at an elevated temperature into, *e.g.*, a fixed bed reactor or other suitable reactor providing intimate contact between the bio-1-alcohol and the catalyst. The fixed bed may be packed with or otherwise arranged to provide sufficient contact with a catalyst. Other suitable reactors may be provided for this purpose provided they can achieve suitable contact for assisting in dehydration. Examples include tubular reactors, fixed and fluidized bed reactors, and the like.

[0074] The reactor can be designed as a simple heated reaction tube packed with catalyst and a flow of the gaseous bio-1-alcohol over the catalyst, preferably in a single pass affording greater than about 95% conversion, with greater than about 98% a preferred embodiment, and in all cases with no recycle stream used whatsoever in the overall process flow. In one

embodiment, the reaction tube is heated isothermally, the tube having from 1 to 10 reaction zone temperatures along the length of the tube, each heated in the range of about 240°C to about 360°C in order to optimize catalyst performance. In a preferred embodiment, the reaction tube has 1 to 4 reaction zone temperatures in the range of about 260°C to about 340°C.

[0075] In one embodiment the process is carried out in a fluidized bed reactor operated in the temperature range of about 240°C to about 360°C. Other continuous flow reactor designs can be used in accordance with procedures and practices used by those familiar with the art of chemical engineering.

[0076] The catalyst used may be prepared by treating γ-alumina (*e.g.*, Porocel® CatGuard®) with at least one base, preferably a promoter, such as with sodium hydroxide, potassium hydroxide, calcium acetate and the like to create a second catalyst. This second catalyst is then calcined to create a third catalyst. The calcination temperature has been found to be most unexpected and critical to creating the most effective catalyst. As seen below when calcined at about 500°C, a significant drop occurs in regioselectivity compared to catalysts calcined at lower temperatures. In one embodiment, as demonstrated in Table 1 below, preferred calcination temperature is about 400°C to about 480°C, and a more preferred calcination temperature is about 440°C to 460°C.

Table 1

Catalyst	Calcination Temperature (°C)	1-Octene (%)	2-Octene & 3-Octenes (%)	1-Octanol (%)	1-Octene Selectivity (%)
EXAMPLE 1	440	94.0	3.2	1.6	97.4
EXAMPLE 2	500	88.9	6.6	3.5	93.1

[0077] Varying catalysts may be used for assisting the bio-1-alcohol in the dehydration step. However, it is preferred that a suitable catalyst that can achieve high levels as regioselectivity as described herein is used. Suitable catalysts include γ- alumina-based catalysts and $ZnAl_2O_4$. Preferably, the catalyst is treated with a base, such as with sodium hydroxide, potassium hydroxide, calcium acetate and the like. In one preferred example, the catalyst is a γ-alumina catalyst treated with a base including a Group I or Group II metal. In

one more preferred embodiment, calcium may be included in the base, and a calcium promoter used. After being treated with the calcium promoter to form a modified γ-alumina catalyst, in a further preferred embodiment, the modified γ-alumina catalyst is then calcined using varying calcination techniques but at temperatures which are preferably much lower than used in standard calcination methods to provide a calcined γ-alumina catalyst. The initial treatment provides a modified γ-alumina catalyst that is then calcined to be ready for use for varying processes including in the dehydration step of the processes herein for making bio-1-alkenes and their subsequent conversion to bio-1,2-alkandiols.

[0078] When employing a calcium promoter, it may be used in the treatment noted above in amounts of about 0.01 weight percent to about 4 weight percent, based on the amount of CaO determined after calcination. In preferred embodiments, about 1 weight percent to about 2 weight percent of the calcium promoter is used based on the amount of CaO determined after calcination.

[0079] In another embodiment, if desired, the alumina-based treated catalyst may be further organosilanized with an organosilane by treatment with, for example, diethoxydiphenylsilane to form an organosilanized base-treated γ-alumina catalyst. The organosilanzized base may be, for example, diethoxydiphenylsilane, dichlorodiphenylsilane and similar materials.

[0080] In one embodiment a catalyst support may comprise $ZnAl_2O_4$ and may also be modified by a base, e.g., by reaction with a promoter and similar catalysts and modified catalyst materials known or to be developed that are capable of carrying out the process as noted herein in a manner similar to the catalysts noted herein that can create a high level of regioselectivity could also be used.

[0081] The catalysts as noted above in addition to being suitable if treated with a base, may alternatively or also be treated with an organosilane or organosilane-modified material to create a promoter-modified catalyst. This promoted catalyst may be further organosilanized with an organosilane by treatment with diethoxydiphenylsilane to create a third, organosilanized and promoter-modified γ-alumina catalyst. The organosilane used to modify the catalyst may be, for example, diethoxydiphenylsilane, dichlorodiphenylsilane and similar materials. As seen in EXAMPLE 10 herein, silanization of the catalyst leads to a decrease in bio-1-octene selectivity, once again showing a difference from previous art and the direct and proven differences in the dehydration catalyst makeup necessary for a highly regioselective dehydration of 1-butanol or 1-octanol.

[0082] In other treatment techniques herein, the γ-alumina catalyst may be modified by the technique of incipient wetting impregnation. A solution of the base is used to modify a solid support in a manner so as to avoid excess moisture that can result in support damage and chemical change. In one embodiment, the incipient wetting impregnation solution (IWS) is about 40% to about 100% the weight of the solid support, which may be, for example an untreated γ-alumina catalyst. In one embodiment, the untreated γ-alumina catalyst is selected from the commercially available, Porocel® CatGuard®. Preferably, the IWS is about 50% to about 60% of the catalyst support weight.

[0083] The γ-alumina may also be provided in a powder form having a surface area of about 50 to about 400 m^2/g. The γ-alumina in one embodiment is shaped by a suitable extrusion process, as are known to those skilled in this art of solid phase catalysts. In one example, the γ-alumina is in the form of 1/16" to 1/8" cylindrical rods with a length to diameter ratio of about 20:1, preferably about a ratio of about 5:1. The extrudate can be in the shape of spheres, star-shaped, hollow cylinders or any three-dimensional shape that can ultimately deliver a γ-alumina with sufficient surface area pore volume, mechanical strength, and physical size to be used in a continuous flow reactor system.

[0084] Once prepared, *e.g.*, a base-treated, preferably a promoter treated, γ-alumina catalyst, as noted above is preferably calcined with care taken in regulating the temperature. This is done at an elevated temperature, preferably in a suitable chamber such as an oven, and the temperature range, while elevated to heat the materials, is not as high as calcination treatments typically used in prior catalyst processes. Instead, the temperature for calcination is kept at about 400°C to about 500°C, and preferably about 420°C to about 480°C, and most preferably about 440°C to about 460°C. Typically, prior standard calcination treatments occur at temperatures 500°C or higher with no limits established or specified for effectiveness. However, applicants discovered that by controlling this temperature and in the specific catalyst treatment process noted herein, the regioselectivity can be unexpectedly controlled to disproportionately favor formation of the bio-1-alkene when the bio-1-alcohol is dehydrated using the resulting calcined catalyst derived from a combination of catalyst treatment and a carefully controlled calcination process.

[0085] In the dehydration step herein, the bio-1-alcohol may be fed to a reactor undiluted or in water through the reactor. Optional an inert gas (*e.g.*, nitrogen) may be used as a purge gas to retain consistency and avoid contaminates in the process. The bio-1-alcohol is preferably

fed, such as by a pump (such as an HPLC pump) or pressurized gas source at a temperature of about 200°C to about 400°C, although temperatures may be adjusted for different bio-1-alcohols depending on the reaction time and flow rate, the catalyst selected and the desired outcome. As the product is removed from the reactor, it is preferably monitored to determine content, identification of product and the ratio of bio-1-alkene conversion as well as the regioselectivity. In a preferred embodiment, the bio-1-alkene has a regioselectivity of at least about 80%, and more preferably about 92% to about 99% and even more preferably about 95% to about 99%.

[0086] In one embodiment, the dehydration process as described in Fig. 1 using the invention catalyst, i.e., the catalyst resulting from the preferred treatment and calcination process noted above, can be stopped and the catalyst maintained under inert gas flow for extended periods of time. This shutdown period can be hours, days or weeks, with heating or no heating applied during this period. The invention catalyst can be re-heated and dehydration process resumed without any detrimental effect on catalyst activity or regioselectivity. Thus, the invention catalysts are not only robust in continuous operation, but they can withstand periods of rest (inactivity) without negative effect on catalyst performance when brought back on line.

[0087] The bio-1-alkene may also be formed using a process in which the bio-1-alcohol in water is dehydrated by heating the bio-1-alcohol with a catalyst under a purge gas as noted above in a bulk process or other continuous reactor. Base and optional organosilane treatments may also be used as well as catalysts and reaction conditions such as those noted above, and most preferably, through use of a calcined γ-alumina catalyst formed by the process described above.

[0088] The bio-1-alcohol may also be formed using a process in which the bio-1-alcohol in water is dehydrated by heating the bio-1-alcohol with a catalyst under a purge, but as noted above, in bulk process or some other continuous reactor. Base and optional organosilane treatments may also be used as well as catalysts and reaction conditions as noted above.

[0089] Once the bio-1-alcohol is dehydrated to form the highly regioselective terminal, bio-1-alkene, that material is then converted to a bio-1,2-alkandiol having a carbon chain length of about 5 to about 20 carbon atoms. Prior to conversion, the bio-1-alkene may optionally be further refined such as by a distillation, vacuum distillation, fractionation or similar refining step to further purify the bio-alkenes to remove trace impurities and minor undesired fractions.

For example, in forming a bio-1-octene, a C8 olefin content of about 90 weight percent to about 99 weight percent can be achieved, and at least about 95 weight percent and preferably about 97 weight percent or greater C8 olefin in the form of the *n*-α-olefin (*i.e.*, 1-octene) can be achieved. In addition, in forming a bio-1-hexene, a C6 content of about 95 weight percent to about 99.8 weight percent can be achieved, with at least about 96 weight percent to about 99 weight percent thereof having a 1-hexene structure.

[0090] Various chemical reactions may be used for this conversion as are known for use in standard alkene to alkanediol conversions using petrochemically derived starting materials and/or that are used for smaller chain bio-alkanes. For example, the bio-1-alkene is reacted in the presence of at least one of formic acid or acetic acid, and a peroxide such as hydrogen peroxide to form an intermediate solution including a bio-1,2-epoxyalkane, bio-1,2-alkanediol, and other components. This is then further contacted with a base solution, such as a sodium hydroxide or another suitable base solution, to complete formation of the bio-1,2-alkanediol. The process noted above preferably yields about 60% to about 99%, and more preferably about 70% to about 99% or about 75% to about 99% of bio-1,2-alkanediol.

[0091] A bio-1-alkanol may also be converted to bio-1-alkene for use in this process by dehydrating the bio-1-alkanol using heat and an acid catalyst in accordance with other previous techniques.

[0092] Bio-1,2-alkanediols, having a chain length of about 5 to about 20 carbon atoms and synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%, more preferably 92% to about 99% and most preferably about 95% to about 99%. In one embodiment, a bio-1-alkanol having a carbon chain of about 5 to about 20 carbon atoms is preferably first converted to a bio-alkene regioselectively such that the resulting bio-alkene contains a bio-1-alkene content of at least about 80% or higher as noted above. Then the bio-1-alkene is converted to the 1,2-alkanediol by any known method known in the art. The resulting bio-1,2-alkanediol after conversion may be further refined such as by distillation, vacuum distillation, fractionation or other similar steps as are noted above for refining the bio-1-alkene to provide a refined, final bio-1,2-alkanediol.

[0093] Resulting bio-1,2-alkanediols, having a chain length of about 5 to about 20 carbon atoms are preferably synthesized by first deyhydrating a bio-1-alkanol having a carbon chain of about 5 to about 20 carbon atoms to a bio-1-alkene having a regioselectivity of at least about

80%, more preferably about 92% to about 99% and most preferably about 95% to about 99% of bio-1-alkene is formed, then treating the bio-1-alkene with formic or acetic acid in the presence of hydrogen peroxide, and further with an aqueous base solution. The bio-1,2-alkanediol may then be separated from the aqueous solution by any suitable technique known in the art of making dihydroxyalkanes in general, and more specifically 1,2-alkanediols. In a preferred embodiment, a distillation step is included for this purpose.

[0094] The bio-1,2-alkanediols as described above, may be used in a variety of compositions, whether used alone or in combination with other different bio-1,2-alkandiols according to the present invention. Such compositions may incorporate one, two or more different bio-1,2-alkanediols according to the invention herein that may act within an antimicrobial system. Such bio-1,2-alkanediol(s), may be used alone as an antimicrobial additive or in an antimicrobial system incorporating other bio-compounds (*i.e.*, any suitable naturally derived, and preferably authentic natural product as tested using mass spectroscopy, gas chromatography and/or the ASTM standard noted above) such as bio-organic acids, bio-diols such as 1,3-propanediol, bio-1,2-butanediol, and 1,2-pentanediol that are extracts from fermentation products of bio-based starting materials and/or other antimicrobial materials, alternative preservatives, traditional preservatives or hurdle technology components as are known in the art or to be developed. An example of a preferred source of bio-compounds for use within the scope of the invention are trigylcerides. In yet a further example, another chemical group of bio-compounds that can be used in preparing alternative preservatives is commonly described as terpenoids. These natural terpenoids can have from 5 to 20 carbon atoms and can be readily obtained through sustainable farming practices, and may be used within the scope of "bio-compounds" as that term is used herein for us in combination with the bio-1,2-alkanediols herein.

[0095] When used in a composition, the bio-1,2-alkanediols of the invention are preferably incorporated in an amount of about 0.1 to about 10 weight percent, and preferably 0.3 to about 2 weight percent, of the total composition; which amount may be varied depending on whether they are used alone for antimicrobial effect or with other components in an antimicrobial system. In the latter instance, the bio-1,2-alkanediols are preferably present in a ratio of the bio-1,2, alkanediol(s) to any other antimicrobial component(s) of about 99:1 to about 1:99, and preferably about 75:25 to about 25:75 in the antimicrobial system (wherein the system would

include the bio-1,2-alkanediol(s) herein and/or any other antimicrobial, traditional preservative, alternative preservation and/or hurdle technology component(s) in the composition).

[0096] When used in certain compositions that are themselves viewed as "antimicrobial products" for providing to other formulators in various industries, the 1,2-bio-alkanediol(s) of the invention may be prepared initially as antimicrobial products that include one or more of the 1,2-bio-alkanediol materials made in accordance with the invention as described above and that incorporate one or more of the compositions as noted above, wherein such antimicrobial products may include also other bio-compounds, known antimicrobials, preservatives, alternative preservation materials or hurdle technology materials.

[0097] Examples of known preservatives and alternative preservative materials or compounds used in alternative preservation systems that may be used with the bio-1,2-alkanediols of the invention, include those suitable for use by various industries in which the present invention may be beneficial. For example, in the cosmetic and personal care industry, the bio-1,2-alkanediol(s) formed according to the present invention can be used alone, in combinations of two or more such materials and/or with other biocompounds and/or with known cosmetic preservatives and alternative preservation materials and/or hurdle technology components. Examples of preservative materials are listed by the FDA in the U.S. as including: (i) traditional preservatives, such as parabens (methyl, ethyl, propyl, and butyl), Quaternium 15 (aka "Dowicil"), diazolidinyl urea, imidazolidinyl urea, DMDM hydantoin, 2-bromo-2-nitropropane-1,3-diol (aka "Bronopol"), sodium hydroxyglycinate, phenoxyethanol, sorbic acid, potassium sorbate, methylisothiazolinone (aka "MI"), methylchloroisothiazoline (aka "CMI" often in combination with MI as Kathon CG), sodium benzoate, caprylyl glycol, sodium dehydroacetate, and formaldehyde; (ii) non-traditional or alternative preservatives: such as extracts of botanicals, organic acids, alcohols and glycerols, fermentation products, glyceryl caprylate, levulinic acid, p-anisic acid, eucalyptus globulus, glycyrrhiza, glabra (licorice) root extract, salvia officinalis, citrus grandis (organic grapefruit) extract, arnica montana (organic arnica) extract, boraxitrus seed extracts, leuconostoc/radish root ferment filtrate. goldseal (hydrastis canadensis root extract), citrus medica, limonum (Lemon) peel extract, caprylhydroxamic acid; and (iii) so-called "self-preserving" materials such as ethanol (when present at > 15%), butylene glycol (when present at >10%), propylene glycol (when present at > 20%); as well as (iv) other components used in alternative or traditional preservation systems in cosmetics such as MDM hydantoin, sodium hydroxymethylglycinate, benzisothiazolinone,

benzyl alcohol, dehydroacetic acid, benzoic acid, salicylic acid, iodopropynyl butylcarbamate chloroxylenol, methyldibromo glutaronitrile, chlorphenesin, triclosan, benzalkonium chloride, chlorhexidine, polyaminopropyl biguanide, 5-bromo-5-nitro-1,3-dioxane (bronidox), hexamidine diisethionate, pentylene glycol, ethylhexylglycerin, triclocarban, glyceryl caprylate, o-cymen-5-ol, chlorphenesin, and glyceryl monolaurate, and traditional petrochemically-derived 1,2-alkanediols such as 1,2-hexanediol, 1,2-octanediol.

[0098] Other countries have similar lists with variations on the nature and type of preservatives, alternative preservatives or preservative system or hurdle technology additives that may be used in those countries. However, the present bio-1,2-alkanediols may be used in such other materials as well. In each of the above-noted systems and compositions, the preferred bio-alkene used in the invention is a bio-octene and the at least one first bio-1,2-alkanediol is bio-1,2-octanediol.

[0099] Examples of compositions that may benefit from an antimicrobial system incorporating one or more of the bio-1,2-alkanediols formed according to the process noted above and/or in compositions as noted above include personal care compositions, such as hair care, oral care, skin care or cosmetic compositions; household product compositions, such as fabric care or cleaning products; industrial compositions; and pharmaceutical, vitamin, nutraceutical or other health care compositions, any of which could benefit from an antimicrobial system, component or antimicrobial product derived from an authentic, natural, bio-based material.

[0100] The bio-1,2-alkanediols are preferably incorporated into a cosmetic or personal care composition (on a wet-basis or total weight basis) so as to make up about 0.001% by weight to about 25% by weight of the composition, and more preferably about 0.01% by weight to about 10% by weight of the composition. The amount used may vary depending on whether other hurdle technology components or other preservatives are also being used in the composition.

[0101] Such bio-1,2-alkanediols alone, in combinations of two or more different bio-1,2-alkandiols, and/or in antimicrobial systems incorporating the bio-1,2-alkanediol(s) alone or with other bio-compounds and/or other preservative, alternative preservative or hurdle technology components, may also be used in a method to provide an antimicrobial effect to a composition, wherein the compositions may be any of the variety of types of compositions as noted above. It is preferred that the bio-1,2-alkanediols herein provide an antimicrobial effect and efficacy to a composition into which the bio-1,2-alkanediols are incorporated, whether into

a personal care, household, industrial, pharmaceutical, vitamin, nutraceutical or other health care composition. Such an antimicrobial effect may be demonstrated by using a variety of suitable antimicrobial effectiveness tests (AET). Such tests include, for example, a compendial test performed during formulation development and stability testing in parenteral drug products intended as a multi-dose product. Suitable test procedures and acceptance criteria are described in the *U.S. Pharmacopeia*, AET, the *European Pharmacopeia* (Efficacy of Antimicrobial Preservation), and the *Japanese Pharmacopeia* (Preservation Effectiveness Tests). Other suitable challenge tests may be used. The FDA also recommends suitable challenge testing.

[0102] One group of antimicrobials, some of which are in the group noted above, are other alcohols, and preferably at least one other alcohol, preferably at least one other diol, and most preferably one or more other vicinal diols that may be petrochemically derived, albeit, it is preferred that such materials are used a diluents or in smaller amounts for providing some antimicrobial effect, but are not so great as to diminish the impact of the natural, bio-based 1,2-alkane diols of the present invention. By the term, "vicinal diols," it is meant materials that have hydroxyl groups which are bonded to atoms in the molecule which are next to each other, *i.e.*, wherein two atoms each bearing a hydroxyl group are bonded to each other. Examples of vicinal diol compounds suitable for use in the invention, include, but are not limited to, ethylene glycol and propylene glycol. Such materials are used in the personal care, cosmetic and pharmaceutical arts as humectants and solvents, and as described in U.S. Publication No. 2007-0207105-A1 as having some modest antimicrobial activity.

[0103] Preferred vicinal diols for use with the bio-1,2-alkanediols herein, in compositions described for use in personal care and pharmaceutical use compositions are medium-chain length, linear vicinal diols that demonstrate some antimicrobial activity including petrochemically-derived 1,2-pentanediol, 1,2-hexanediol, 1,2-octanediol, and 1,2-decanediol. Other vicinal diols useful in the compositions described herein include molecules derived from glycerin. Glycerin can be reacted with other molecules at its 1- or 3-position, leaving two vicinal hydroxyl groups. For example, glyceryl monoethers, such as ethylhexylglycerin [3-(2-ethylhexyloxy)propane-1,2-diol], available commercially as SENSIVA®SC50 from Schulke & Mayr, are useful, traditional, liquid vicinal diols having antimicrobial properties. Glyceryl monoesters such as glyceryl monolaurate, glyceryl mono caproate, or glyceryl monocaprylate, the latter of which is commercially available from Inolex Chemical Company, Philadelphia, PA, are also useful antimicrobial vicinal diols. For the preservation of cosmetics, toiletries and

pharmaceuticals, vicinal diols are known to be effective against bacteria and yeast but weak against fungi and to date there are only limited natural choices, none of which to applicant's knowledge are bio-derived natural 1,2-alkanediols of from 5 to 20 carbon atoms.

[0104] The compositions herein preferably do not include, or include only to a minor amount any preservation materials such as parabens, or other known preservation materials that can be viewed as harmful to the user.

[0105] Such personal care and pharmaceutical compositions may optionally further include a solubilizing agent in amounts of about 1% to about 70% by weight of the combination of the solubilizing material with the bio-1,2-alkane diol(s) (or if used with other preservation component(s) by weight of the entire antimicrobial system with the solubilizing material). Suitable solubilizing agents include vicinal and other traditional diols.

[0106] Also useful in with the bio-1,2-alkanediols in any antimicrobial system are hydroxamic acid and the like which may be used in a variety of industries. Suitable hydroxamic acids include alkylhydroxamic acids and bio-alkylhydroxamic acids that include at least one alkyl group of a chain length of about two to about twenty-two carbon atoms, which may be branched or linear in structure, substituted or unsubstituted, and saturated or unsaturated as noted hereinabove. Preferred alkylhydroxamic acids contain alkyl groups of a chain length of about six to about twelve carbon atoms and most preferably linear chains of that length. Most preferred alkylhydroxamic acids are caprylohydroxamic acid, having a linear terminal chain of eight carbon atoms and caprohydroxamic acid, having a linear chain of ten carbon atoms. Such alkylhydroxamic acids may be used alone or in combination and in amounts as noted above for an optional solubilizing agent.

[0107] Formulations prepared for personal care use, and pharmaceutical compositions, depending on the end use (topical or oral) may include any other colorants, fragrances, active ingredients or other additives typically used and/or to be developed in the art for use in personal care and pharmaceutical formulations, in which additives will vary depending upon the formulation in which the preferred bio-1,2-alkanediols, or antimicrobial products or systems containing the bio-1,2-alkanediols are used, for example, in topical skin toners, skin cleansers, night creams, skin creams, shaving creams, skin care lotions, or other cosmetic preparations; make-up, such as foundation, liquid and powder-based make-up, mascara, lipstick, blush, gloss, eye-liner and the like; or other personal care and/or pharmaceutical compositions, such as, sunscreens, lip balms, fragrances, massage oil, shampoos, conditioners, conditioning shampoos,

hair styling gels, hair reparatives, hair tonics, hair fixatives, hair mousses, bath and shower gels, liquid soaps, moisturizing sprays, makeup, pressed powder formulations, bath additives, ophthalmic preparations, foaming soaps and body washes, sanitizing wipes, hand sanitizers, medicaments (tablets or liquid), towelettes and wipes and others. It should be understood, based on this disclosure that a wide variety of personal care and pharmaceutical compositions could benefit from the properties of the bio-1,2-alkanediols of the present invention, wherein, as used herein, pharmaceutical composition is one having at least one active pharmaceutical ingredient (API).

[0108] The personal care and pharmaceutical formulations, if liquid based (such as gels, hydrogels, lotions, shampoos, liquid pharmaceuticals and the like) will also preferably include water as part of the liquid base. The formulations and compositions may include other additives as well depending on the end formulation desired, such as without limitation, at least one humectant, at least one emulsifier and/or thickener, chelating agent(s), gelling agent(s), amino acid(s), emollient(s), various solvents, free radicals and initiators, sunscreen UVA and/or UVB blocking agents, antioxidants, other preservatives, waxes, polymers and copolymers, inorganic and organic pigments and/or one more fragrances, favoring(s), coloring agent(s), herbs, natural extracts, essential oils, pharmaceutical drug products, other APIs and other additives commonly used in such formulations.

[0109] Personal care and pharmaceutical compositions using the bio-1,2-alkanediols herein may be lotion-based, oil-in-water emulsions, water-in-oil emulsions, water-in-silicone emulsions, silicon-in-water emulsions, gels, solids, liquids, cream based, oil based, aqueous/alcoholic or glycolic solution based, dispersions, suspensions or syrups, microemulsions or a liposome-based formulations.

[0110] In water-based formulations, other than solids and thicker gels, etc., it is preferred that about 20% by weight to about 95% by weight (on a wet basis) of water is incorporated therein. The various additives aside from the water and preferred antimicrobial systems noted herein, would make up the remaining portion of various personal care and pharmaceutical compositions. Preferably, each additive is present in an amount of up to about 75 percent by weight of the entire formulation, and more preferably up to about 40 percent by weight, with a collective amount of such additives of preferably no greater than about 50 percent by weight.

[0111] In a household product, antimicrobials are also useful additives. Household product compositions can include household cleaners as well as fabric care compositions. Cleaning

compositions (whether solids or solutions) can incorporate cleaning agents that are active ingredients for cleaning (bleach, vinegar, ammonia, citric acid and the like). For liquid cleaning agents, aqueous solutions may be used as are known in the art or to be developed, such as quaternary ammonium compounds, bleach, vinegar, or basic or acidic cleansers. Commercially available quaternary ammonium-based cleaning products include various antibacterial all-purpose cleaners among others which are intended and well-suited for sanitizing or disinfecting efficacy, however, such compositions still incorporate antimicrobials and other preservatives for shelf life and to prevent growth of foreign agents. Thus, such compositions also benefit from use of the natural bio-1,2-alkanediols herein.

[0112] For more natural cleaning solutions, such as citric acid-based agents or other green cleaning agents (ecologically friendly), consumers would be interested in further additives that are also natural, and in this case that are derived from a biological source. Thus, such compositions benefit from natural bio-based 1,2-alkanediols herein as well as antimicrobial systems and products incorporating the bio-1,2-alkanediols. Such natural cleaning compositions as well as standard cleaning compositions as noted above may also incorporate various optional additives in varying amounts as noted below. Citric acid-based cleaning products may include lemon, orange or grapefruit-based cleaning agents. Other suitable components may include grape seed oil, vegetable oils combined with one or more of mild peroxide agents, surfactants, and the like, as well as pigment(s) or colorant additive(s) to provide a visual alerts when the cleaners are present, conventional or alternative preservation compound(s), antimicrobial(s), bactericide(s) or fungicide(s) (each of which may be used along or in combination with the bio-1,2-alkanediols of the present invention in an antimicrobial system or product), thixotropic agents and rheological modifiers, pH adjusting additives or buffers, as well as fragrance additive(s) for providing a clean smell (pine scent, lemon scent, orange scent, floral scent, etc.). In addition, other agents for foaming, color change or effervescence (bubbling) may be provided if desired to demonstrate cleaning action. Solid cleaners can incorporate similar additives, but can be compressed, incorporated or formed from depositing a cleaning material on a substrate (sponge, scrubber, mop head or the like), and various inactive agents for holding the solid cleaning material together (hardeners, gelling agents, mineral powder and the like). Other additives for household cleaning compositions may include pH buffering agent(s), a fragrance encapsulate(s) or carrier(s), fluorescer(s), hydrotrope(s), soil-release agent(s), polyelectrolyte(s), enzyme(s), optical brightening agent(s),

anti-oxidant(s), UV absorbing compound(s), propylene glycol, dipropylene glycol, opacifier(s), pearlescent(s) and combinations of these as well as other cleaning product additives known or to be developed.

[0113] The fabric care compositions can include any active fabric cleaning component as well as various conditioning components as are known or to be developed in the art. Such compositions may include ingredients used for fabric cleaning or laundering, fabric conditioning or softening, fabric dye(s), water conditioner(s) and/or fabric care composition(s) such as spot removers or stain treatments. Such ingredients include without limitation antifoam agent(s), antideposition agent(s), fragrance(s) and their carrier(s) or encapsulate(s), traditional or conventional diols (although such diols may be omitted if the bio-1,2-alkanediols are useful in the same or an enhanced or boosted capacity for an existing diol), colorant(s) such as pigment(s) or dye(s), and co-softening agent(s). Others additives for fabric care compositions may include, without limitation, other traditional preservative(s), antimicrobial(s), bacteriocide(s), and/or fungicide(s)(each of which may be used alone or in an antimicrobial product or system as described herein), pH modifier(s) or buffering agent(s), fluorescer(s), hydrotrope(s), soil-release agent(s), polyelectrolyte(s), enzyme(s), optical brightening agent(s), anti-shrinking agent(s), anti-wrinkle agent(s), anti-spotting agent(s), anti-oxidant(s), UV absorbing compound(s), anti-corrosion agent(s), drape imparting agent(s), anti-static agent(s), ironing aid(s), odor-preventing compound(s), perfume encapsulate(s), cotton seed oil, tea tree oil, aloe vera extract, propylene glycol, dipropylene glycol, opacifier(s), pearlescent(s) and combinations of these components.

[0114] The antimicrobial bio-1,2-alkanediols herein may also be used in the treatment of the finished textile(s) and/or treatment of the fibers or yarns from which a textile is to be fabricated to achieve the desired benefit. Textiles may include woven and non-woven textiles, such as felt and tapa cloth and other bark cloths, or blends and combinations of the same. Textile fiber (whether it exists in finished textile form, yarn or fiber form) may be any known or developed in the art and include synthetic fibers, "natural" fibers, such as animal derived fibers or cellulosic/plant-derived fibers and blends or combinations of any of these.

[0115] Animal-derived fibers may include and derived from the hairs or fur of an animal. Examples are, without limitation, lambs or sheep's wool, alpaca, angora wool, azlon, byssus, camel hair, cashmere wool, chiengora, chatgora, llama, mohair wool, qiviut, rabbit, silk, vicuna, yak, pashmina wool and combinations of the same. Cellulosic or plant-derived fibers may

include, without limitation, those obtained from flax (linen fibers), cotton, ramie, jute, kenaf, beach hibiscus, roselle, urena, hemp (e.g., Crotalaria juncea, Cannabis sativa, Apocynum cannabinum), hoop vine, sisal, henequen, yucca, abaca, genus Sansevieria, New Zealand flax, cotton, coir, milkweed, kapok, floss silk, Proboscidea parviflora, bamboo, bast, fique, banana, modal, lyocell, pina, raffia, rayon, soy protein, acetate and combinations of the same. Synthetic fibers may include any known or to be developed, such as for example, and without limitation, acrylic, Kevlar®, modacrylic, Nomex®, nylon, polyester, lycra, spandex, rayon, and combinations of the same.

[0116] Fabric treatment compositions may be applied to the textile or fibers. The compositions may be applied as either a wet or dry composition and during a laundering cycle with water or a drying cycle. Such fabric treatment materials may be used for softening and/or conditioning a textile or fiber; reducing and/or preventing wrinkles, imparting fragrances to a textile or fiber; reducing ironing time, improving softness and the like.

[0117] The invention also incorporates use of the 1,2-bio-alkanediol materials formed according to the invention as a material for boosting the antimicrobial efficiency of existing antimicrobials, preservatives, alternative preservation materials and/or hurdle technologies.

[0118] By boosting the antimicrobial efficacy, it is meant that the antimicrobial efficacy of an existing antimicrobial and/or preservative (including alternative preservation materials or hurdle technologies) when combined with the 1,2-bio-alkandiols achieve an improvement of antimicrobial effectiveness that is more than insubstantial, *i.e.*, at least about 5%, preferably at least about 10%, and more preferably at least about 20% or more improvement, while providing a natural boosting material. Boosting effects and antimicrobial effects in a formulation are evaluated by measuring performance using a preservative efficacy test ("PET") or a challenge test. Preservatives and antimicrobials used in cosmetic, toiletry and pharmaceutical products must enable the products to successfully pass microbiological testing protocols, known as "challenge tests," established by government regulations and trade organizations. Challenge tests are performed by adding known quantities of microorganisms to a product and measuring the increase or decrease in microorganism population over time. The organisms include Gram-positive bacteria, Gram-negative bacteria, yeast and mold. The Cosmetic, Toiletries, and Fragrance Association (CTFA) has defined a challenge test that is widely accepted as the standard in the cosmetic, toiletry and pharmaceutical industry. The test requires that the quantity of bacteria be reduced by 99% in seven days, and that the quantity of yeast and fungi

(mold) be reduced by 90% in seven days. In order to pass a challenge test, the product must contain the appropriate amounts and types of preservative compounds that will enable antimicrobial efficacy against a broad spectrum of microorganisms in a short period of time.

[0119] In addition to the content noted to provide antimicrobial efficacy, if using as a boosting material to improve the effectiveness of an existing antimicrobial and the like as noted above it is preferred that the 1,2-bioalkanediol(s) are incorporated in an amount of about 0.1 to about 10 weight percent, and preferably 5.0 to about 3.0 weight percent, of the total composition; and in a ratio of about 99.999:0.001 to about 0.001:99.999, and preferably about 99.9:0.1 to about 0.1:99.9 of the total amount of existing antimicrobial, preservative, alternative preservation and/or hurdle technology components already existing in the composition.

[0120] The invention will now be described with respect to the following non-limiting examples:

EXAMPLES

[0121] Bio-1-octanol is formed from the hydrogenation of bio-octanoic acid (suitable sources of which may be derived from coconut, palm or any other renewable source or process that can produce octanoic acid). The bio-1-octanol is dehydrated regioselectively by feeding the bio-1-octanol at an elevated temperature into a fixed bed reactor containing a catalyst according to the invention herein.

[0122] Gas Chromatography (GC) was an important method used to characterize both petroleum and natural-renewable derivatives. Products (*e.g.* octenes) were characterized using a Thermo Scientific Trace 1310 Gas Chromatograph employing an FID detector and Chromeleon software (version 7.2.4.8525). For dehydration reactions products the GC employed a Restek MXT-5 column (30 m length, 0.5 μm film, 0.53 ID), helium carry gas (3.0 mL/min), CT split injection 45 mL/min (5.0 mL/min purge flow), injector 250 °C, detector 300 °C, oven 70 °C hold for 8 min, then ramp to 300 °C (15 °C/min) with a final hold for 6.67 min. For alkanediol products GC analysis employed a Restek MXT-WAX column (30 m length, 0.5 μm film, 0.53 ID), helium carry gas (5.0 mL/min), CT split injection 10 mL/min (5.0 mL/min purge flow), injector 220 °C, detector 250 °C, oven 70 °C hold for 0.25 min, then ramp to 250 °C (10 °C/min) with a final hold for 6.67 min.

EXAMPLE 1

[0123] γ-Alumina powder was treated with an aqueous solution of calcium acetate using the technique of incipient wetting to afford a catalyst containing 1.50% calcium (based on CaO) after calcination in air at 440°C for a period of 12 h.

EXAMPLE 2

[0124] γ-alumina powder was treated with an aqueous solution of calcium acetate using the technique of incipient wetting to afford a catalyst containing 1.50% calcium (based on CaO) after calcination in air at 500°C for a period of 12 h.

EXAMPLE 3

[0125] Catalysts prepared in EXAMPLE 1 and EXAMPLE 2 were loaded into separate 75 mL (Sigma Aldrich Part No. Z173592) reactors, each was wrapped with heating tape and covered with fiberglass insulation. The temperature was controlled by affixing a thermocouple to the outside skin of the reactor, this input was used for a temperature controller. Bio-1-octanol was fed into the reactor maintained at 315°C using an HPLC pump. The bio-1-octanol feed rate was adjusted in a manner to obtain conversion more than 95%, but less than 100%. After 1 h of continuous operation a sample was collected and analyzed for analyzing chemical conversion of the bio-1-octanol and regioselectivity of bio-1-octene. The results are shown below:

Table 2

Catalyst	Calcination Temperature (°C)	1-Octene (%)	2-Octene & 3-Octenes (%)	1-Octanol (%)	1-Octene Selectivity (%)
EXAMPLE 1	440	94.0	3.2	1.6	97.4
EXAMPLE 2	500	88.9	6.6	3.5	93.1

EXAMPLE 4

[0126] γ-alumina in the form of 1/16" extrudate, Porocel® CatGuard® was cut to have length to diameter ratios in the range of 2 to 4. The extrudate was modified with an aqueous solution of calcium acetate. The weight of solution to alumina was 0.55 by weight and the calcium acetate concentration was at level to deliver 1.50% of CaO (after calcination). This catalyst was subjected to calcination in air at 440°C for a period of 12 h.

EXAMPLE 5

[0127] The 48 g of the catalyst prepared in EXAMPLE 4 was loaded into a 75 mL (Sigma Aldrich Part No. Z173592) reactor, wrapped with heating tape, and covered with fiberglass insulation. The temperature was controlled by affixing a thermocouple to the outside skin of the reactor, this input was used for a temperature controller. Bio-1-octanol was fed at 0.2 mL/min into the reactor maintained at 315°C. After >1,400 h of TOS, that included regular shut-downs and start-ups, catalyst performance with regard 1) chemical conversion of the bio-1-octanol, 2) product selectivity, 3) and regioselectivity for bio-1-octene was unchanged. A GC chromatogram of the crude product collected from the reactor is shown in Fig. 3 with an octene selectivity of 96% and with chemical yield of bio-1-octene of 94%.

EXAMPLE 6

[0128] The product in EXAMPLE 5 was subjected to simple distillation at atmospheric pressure. The product obtained was analyzed by GC and the chromatogram is shown in Fig. 4. The product from the distillation was 99.9% bio-octenes with a 96.7% content of bio-1-octene.

EXAMPLE 7

[0129] The product in EXAMPLE 6 was converted to the bio-1,2-octanediol by treatment with formic acid and peroxides, then a base hydrolysis, and finally purified by fractional distillation. The GC chromatogram is shown in Fig. 5. The bio-1,2-octanediol obtained in this example has a chemical purity 98.7% by GC analysis. A typical GC chromatogram of the petro-1,2-octanediol currently used in commercial cosmetic applications is displayed in Fig. 6.

EXAMPLE 8

[0130] Stable isotope analyses were carried out on petroleum-based and those prepared from the natural and renewable feedstocks prepared in EXAMPLES 6 and 7. The values are shown below:

Table 3

Compound	$\delta^{13}C$	$\delta^{2}H$
Bio-1-Octanol	32.3	-273
1-Octanol (petro-based)	30.3	-87
Bio-1-Octene	32.0	-288
1-Octene (petro-based)	31.8	Not tested
Bio-1,2-Octanediol	32.2	-245
1,2-Octanediol (petro-based)	31.5	-50

EXAMPLE 9

[0131] The bio-1,2-octanediol prepared in EXAMPLE 7 was subjected to testing that is currently used to qualify the petroleum, non-renewable, non-natural equivalent. The bio-1,2-octanediol prepared in EXAMPLE 7 was found to meet all the batch analytical testing requirements:

Table 4

Analysis	Natural 1,2-octanediol	Petroleum 1,2-octanediol	Specification
Odor	Conforming	conforming	mild
Color, Lovibond (yellow)	0.1	0.1	≤ 1.5
Color, Lovibond (red)	0.0	0.0	≤ 0.3
Sediment	Conforming	Conforming	No discoloration
Color, APHA 142	8	1	≤ 100
COLOR, APHA 142-01	8	1	≤ 30
Infrared Spectrum	Conforming	Conforming	Matches Std.
Hydroxyl value Mg KOH/g	761	740	740 to 770
Moisture % w/w	0	0	≤ 2
Appearance	Conforming	Conforming	Clear liquid above 35 °C
Assay, %	98.7	99.5	≥ 98.0
% 1,2-octane oxide	0.1	0.1	Report result
Color 999-03	Conforming	Conforming	Matches Standard

EXAMPLE 10

[0132] In a comparative example, bio-1-octanol was fed into a 75 mL reactor packed with a catalyst prepared as follows: gamma-alumina powder was modified with a sodium hydroxide solution (5 wt-% NaOH) to create a second catalyst, the second catalyst was dried in an oven and then treated with diethoxydiphenylsilane (1 wt-% based on alumina) dissolved in ethanol. The solvent was removed, the catalyst washed with ethanol, and dried in an oven to create a

third catalyst. This third catalyst was calcined at 400°C for 12 h. The catalyst was loaded into a 75 mL reactor, heated to a target temperature of 325°C and bio-1-octanol feed rates were adjusted to optimize both conversion and regioselectivity.

EXAMPLE 11

[0133] Petro-1-hexanol was fed into a 75 mL reactor packed with the catalyst prepared in EXAMPLE 4. The catalyst used in this example had greater than 1,400 h of TOS used for bio-1-octanol dehydration to bio-1-octene. For this series of dehydration reaction examples, the temperature was varied from 330°C to 370°C and the feed rate was adjusted in order to achieve between 1 and 2% of hexanol passing through the reactor. The results for the hexanol dehydration are shown below:

Table 5

Catalyst	Reactor Temperature (°C)	1-Hexene (%)	2-Hexenes & 3-Hexene (%)	1-Hexanol (%)	1-Hexene Selectivity (%)
EXAMPLE 11A	330	92.8	3.5	1.8	95.8
EXAMPLE 11B	350	93.8	3.1	1.7	96.8
EXAMPLE 11C	370	90.1	3.2	1.6	96.6

[0134] In EXAMPLE 11C where the reactor is maintained at 370°C a new peak in the GC chromatogram appears have an area-% of 3.6; thus, indicating a lower chemical yield of 1-hexene and formation of another product.

EXAMPLE 12

[0135] After 1400 h of TOS for the dehydration of bio-1-octanol, some catalyst was removed at the entrance side of the tube reactor. In Fig. 7 the used catalyst is photographed next to catalyst with 0 h TOS. The used catalyst shows only a slight discoloration, and as noted herein, displays no loss in chemical conversion or loss in regioselectivity.

[0136] It will be appreciated by those skilled in the art that changes could be made to the embodiments described above without departing from the broad inventive concept thereof. It is

understood, therefore, that this invention is not limited to the particular embodiments disclosed, but it is intended to cover modifications within the spirit and scope of the present invention as defined by the appended claims.

CLAIMS

We claim:

1. A process for the synthesis of bio-1,2-alkanediols, comprising:

providing a bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%; and

converting the bio-alkene to a bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms.

2. The process of claim 1, wherein the bio-alkene and the bio-1,2-alkanediol each have a carbon chain of about 6 to about 14 carbon atoms.

3. The process according to claim 2, wherein the bio-alkene and the bio-1,2-alkanediol each have a carbon chain length of about 6 to about 10 carbon atoms.

4. The process according to claim 3, wherein the bio-alkene and the bio-1,2-alkanediol each have a carbon chain length of about 6 to about 8 carbon atoms.

5. The process according to claim 1, wherein the bio-alkene is bio-octene and the bio-1,2-alkane diol is bio-1,2-octanediol.

6. The process according to claim 1, wherein the bio-alkene results from a process wherein a bio-1-alcohol is dehydrated by heating the bio-1-alcohol in a reactor with a catalyst.

7. The process according to claim 6, wherein the reactor is a fixed bed reactor.

8. The process according to claim 7, wherein the fixed bed reactor is a fluidized fixed bed reactor.

9. The process according to claim 6, wherein the catalyst is selected from $ZnAl_2O_4$ and a ɤ-alumina catalyst.

10. The process according to claim 9, wherein the bio-alkene has a bio-1-alkene regioselectivity of about 92% to about 99%.

11. The process according to claim 10, wherein the bio-alkene have a bio-1-alkene regioselectivity of about 95% to about 99%.

12. The process according to claim 1, wherein the process yields at least about 92% to about 99% of the bio-1,2-alkanediol.

13. The process according to claim 9, wherein the catalyst is a γ-alumina catalyst is treated with a base to form a modified γ-alumina catalyst.

14. The process according to claim 13, wherein the base comprises a Group I or a Group II metal.

15. The process according to claim 9, wherein the γ-alumina catalyst is treated with a calcium promoter to provide a modified γ-alumina catalyst.

16. The process according to claim 15, wherein the modified γ-alumina catalyst is calcined to provide a calcined γ-alumina catalyst.

17. The process according to claim 16, wherein the calcium promoter is used in an amount of about 0.01 weight percent to about 4 weight percent based on the weight of CaO determined after calcination.

18. The process according to claim 17, wherein the calcium promoter is used in an amount of about 1 weight percent to about 2 weight percent based on the weight of CaO determined after calcination.

19. The process according to claim 15 wherein a temperature during calcination is about 400°C to about 500°C.

20. The process according to claim 19, wherein the temperature during calcination is about 420°C to about 480°C.

21. The process according to claim 20, wherein the temperature during calcination is about 440°C to about 460°C.

22. The process according to claim 15, wherein calcination is carried out in an oven.

23. The process according to claim 15, wherein the calcination is carried out in an atmosphere of air or inert gas.

24. The process according to claim 6, wherein the bio-alkene undergoes a distillation step to refine the bio-alkene prior to converting the bio-alkene to the bio-1,2-alkanediol.

25. The process according to claim 6, wherein the bio-1,2-alkanediol formed from the bio-alkene undergoes a final distillation step to refine the bio-1,2-alkanediol.

26. The process according to claim 1, wherein the bio-alkene results from a process wherein the bio-1-alcohol in water is dehydrated by heating the bio-1-alcohol with a catalyst under a purge gas.

27. The process according to claim 1, wherein the bio-alkene is converted to the bio-1,2-alkanediol by reacting the bio-alkene in the presence of at least one of formic acid or acetic acid and a peroxide to form a bio-1,2-epoxyalkane having an epoxy ring, and contacting the 1,2-epoxyalkane with water and sodium hydroxide to form the bio-1,2-alkanediol.

28. The process according to claim 1, wherein the process yields at least about 60% to about 99% of the bio-1,2-alkanediol.

29. The process according to claim 28, wherein the process yields at least about 72% to about 99% of the bio-1,2-alkanediol.

30. A process for making bio-1,2-alkanediols, comprising:

providing a bio-1-alcohol and a catalyst treated with a base;

dehydrating the bio-1-alcohol in the presence of the catalyst to form a bioalkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%; and

converting the bio-alkene to a bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms.

31. The process according to claim 30, wherein the catalyst is a γ-alumina catalyst and the base comprises calcium.

32. The process according to claim 31, wherein the γ-alumina catalyst is treated with a calcium promotor and after treatment is calcined at a temperature of about 400°C to about 500°C.

33. The process according to claim 30, wherein the process yields about 72% to about 99% of the bio-1,2-alkanediol.

34. The process according to claim 30, wherein the bio-1-alkene regioselectivity is about 95% to about 99%.

35. A method for treating a catalyst for use in dehydration of alcohols, comprising

providing a γ-alumina catalyst;

treating the γ-alumina catalyst with a base comprising a Group I or a Group II metal; and

heating the γ-alumina catalyst to a temperature of about 400°C to about 500°C.

36. The method according to claim 35, wherein the base comprises calcium.

37. The method according to claim 36, wherein the base is a calcium promoter and the heating occurs during calcination.

38. The method according to claim 37, wherein the calcium promoter includes is used in an amount of about 0.01 weight percent to about 4 weight percent based on the weight of CaO determined after calcination.

39. The process according to claim 38, wherein the calcium promoter is used in an amount of about 1 weight percent to about 2 weight percent based on the weight of CaO determined after calcination.

40. The process according to claim 35, temperature during calcination is about 420°C to about 480°C.

41. The process according to claim 40, wherein the temperature during calcination is about 440°C to about 460°C.

42. A composition, comprising at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

43. The composition according to claim 42, wherein the composition is a personal care composition.

44. The composition according to claim 43, wherein the personal care composition is a hair care composition, an oral care composition, a skin care composition or a cosmetic composition.

45. The composition according to claim 42, wherein the composition is a composition for a household product.

46. The composition according to claim 45, wherein the household product is a fabric care product or a cleaning product.

47. The composition according to claim 42, wherein the composition is an industrial composition.

48. The composition according to claim 42, wherein the composition is a pharmaceutical composition, a vitamin composition, or a health care composition.

49. The composition according to claim 42, comprising a second bio-1,2-alkanediol, different than the first bio-1,2-alkanediol, the second bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a second bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

50. The composition according to claim 49, wherein the composition further comprises at least one other bio-compound different from the first and the second bio-1,2-alkanediols.

51. The composition according to claim 50, wherein the composition comprises an antimicrobial compound different from the first and the second bio-1,2-alkanediol and different from the other bio-compound.

52. The composition according to claim 42, wherein the composition further comprises at least one other bio-compound different from the at least one bio-1,2-alkanediol.

53. The composition according to claim 52, wherein the composition comprises at least one antimicrobial compound different from the at least one bio-1,2-alkanediol and different from the other bio-compound.

54. The composition according to claim 42, wherein the first bio-1-alkene has a regioselectivity of about 95% to about 99%.

55. The composition according to claim 42, wherein the first bio-alkene is bio-octene and the at least one first bio-1,2-alkane diol is bio-1,2-octanediol.

56. A method for providing an antimicrobial effect to a composition, including incorporating an antimicrobial system to the composition, wherein the antimicrobial system comprises at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of at least one first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

57. The method according to claim 56, wherein the antimicrobial system further comprises a second bio-1,2-alkanediol different than the first bio-1,2-alkanediol, wherein the first bio-1,2-alkanediol has a carbon chain length of about 5 to about 20 carbon atoms and is formed by conversion of a second bio-alkene, different than the first bio-alkene, having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

58. The method according to claim 57, wherein the composition further comprises at least one other bio-compound different from the first and the second bio-1,2-alkanediols.

59. The method according to claim 58, wherein the antimicrobial system comprises at least one other preservative compound different from the first and the second bio-1,2-alkanediol and different from the other bio-compound.

60. The method according to claim 57, wherein the composition further comprises at least one other bio-compound different from the at least one first bio-1,2-alkanediol.

61. The method according to claim 60, wherein the antimicrobial comprises at least one other antimicrobial compound and/or preservative compound different from the at least one first bio-1,2-alkanediol and different from the other bio-compound.

62. The method according to claim 57, wherein the antimicrobial system demonstrates antimicrobial efficacy.

63. The method according to claim 57, wherein the first bio-1-alkene has a regioselectivity of about 95% to about 99%.

64. The method according to claim 57, wherein the first bio-alkene is bio-octene and the at least one first 1,2-alkane diol is bio-1,2-octanediol.

65. A method for boosting the antimicrobial efficiency of an antimicrobial and/or a preservative in a composition, comprising incorporating an antimicrobial system into the composition, wherein the second antimicrobial system comprises at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

66. The method according to claim 65, wherein the antimicrobial system further comprises a second bio-1,2-alkanediol different than the first bio-1,2-alkanediol, wherein the second bio-1,2-alkanediol has a carbon chain length of about 5 to about 20 carbon atoms and is formed by conversion of a second bio-alkene, different than the first bio-alkene, having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

67. The method according to claim 66, wherein the composition further comprises at least one other bio-compound different from the antimicrobial and/or preservative and different from the first and the second bio-1,2-alkanediols.

68. The method according to claim 65, wherein the composition further comprises at least one other bio-compound different from the at least one first bio-1,2-alkanediol.

69. The method according to claim 65, wherein the first bio-1-alkene has a regioselectivity of about 95% to about 99%.

70. The method according to claim 65, wherein the antimicrobial system demonstrates antimicrobial efficacy.

71. The method according to claim 65, wherein the first bio-alkene is bio-octene and the at least one first bio-1,2-alkane diol is bio-1,2-octanediol.

72. An antimicrobial product, comprising at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

73. The antimicrobial product according to claim 72, further comprising a second bio-1,2-alkanediol, different than the at least one first bio-1,2-alkanediol, the second bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms and formed by conversion of a second bio-alkene, different than the first bio-alkene, having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

74. The antimicrobial product according to claim 73, wherein the product further comprises at least one other bio-compound different from the at least one first and the second bio-1,2-alkanediols.

75. The antimicrobial product according to claim 74, further comprising a second antimicrobial and/or a preservative different from the antimicrobial product.

76. The antimicrobial product according to claim 72, wherein the product further comprises at least one other bio-compound different from the at least one first bio-1,2-alkanediol.

77. The antimicrobial product according to claim 76, further comprising a second antimicrobial and/or a preservative different from the antimicrobial product.

78. The antimicrobial product according to claim 72, wherein the first bio-1-alkene has a regioselectivity of about 95% to about 99%.

79. The antimicrobial product according to claim 72, wherein the antimicrobial product demonstrates antimicrobial efficacy.

80. The antimicrobial product according to claim 72, wherein the first bio-alkene is bio-octene and the at least one first 1,2-alkane diol is bio-1,2-octanediol.

81. A product for boosting the efficacy of an antimicrobial and/or a preservative in a composition, the product comprising at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

82. The product according to claim 81, further comprising a second bio-1,2-alkanediol, different than the at least one first bio-1,2-alkanediol, the second bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms and formed by conversion of a second

bio-alkene, different than the first bio-alkene, having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

83. The product according to claim 82, wherein the composition further comprises at least one other bio-compound different from the at least one first and the second bio-1,2-alkanediols.

84. The product according to claim 81, wherein the product further comprises at least one other bio-compound different from the at least one first bio-1,2-alkanediol.

85. The product according to claim 81, wherein the first bio-1-alkene has a regioselectivity of about 92% to about 99%.

86. The product according to claim 82, wherein the first bio-alkene is bio-octene and the at least one first bio-1,2-alkane diol is bio-1,2-octanediol.

87. The product according to claim 81, wherein the product demonstrates antimicrobial efficacy.

ABSTRACT OF THE DISCLOSURE

A process is incorporated herein for the synthesis of bio-1,2-alkanediols, comprising: providing a bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%, at least about 92% and/or at least about 95%; and converting the bio-alkene to a bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms. Methods for treating catalysts which may be incorporated in the process for the synthesis of bio-1,2-alkanediols are also included herein. Such bio-1,2-alkanediols are used in compositions and products alone as antimicrobial materials, or with existing bio-compounds and/or antimicrobials, preservatives, alternative preservation systems and/or hurdle technology components. The bio-1,2-alkanediols incorporate a natural and bio-based pathway for antimicrobial effects in various compositions such as cosmetic, pharmaceutical, industrial and household products.

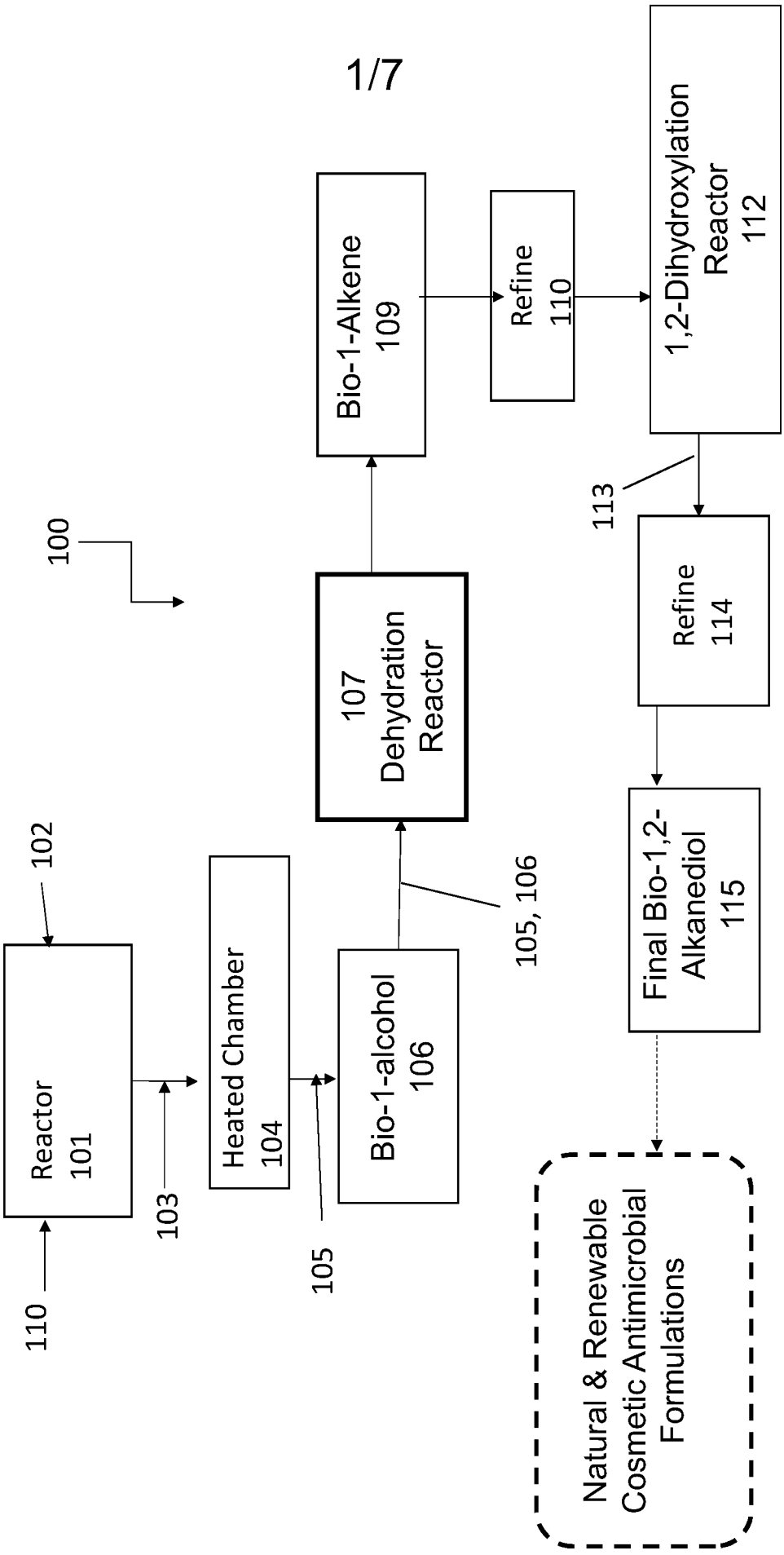

100
Reactor 101
102
110
103
Heated Chamber 104
105
Bio-1-alcohol 106
105, 106
107 Dehydration Reactor
Bio-1-Alkene 109
Refine 110
1,2-Dihydroxylation Reactor 112
113
Refine 114
Final Bio-1,2-Alkanediol 115
Natural & Renewable Cosmetic Antimicrobial Formulations

Fig. 1

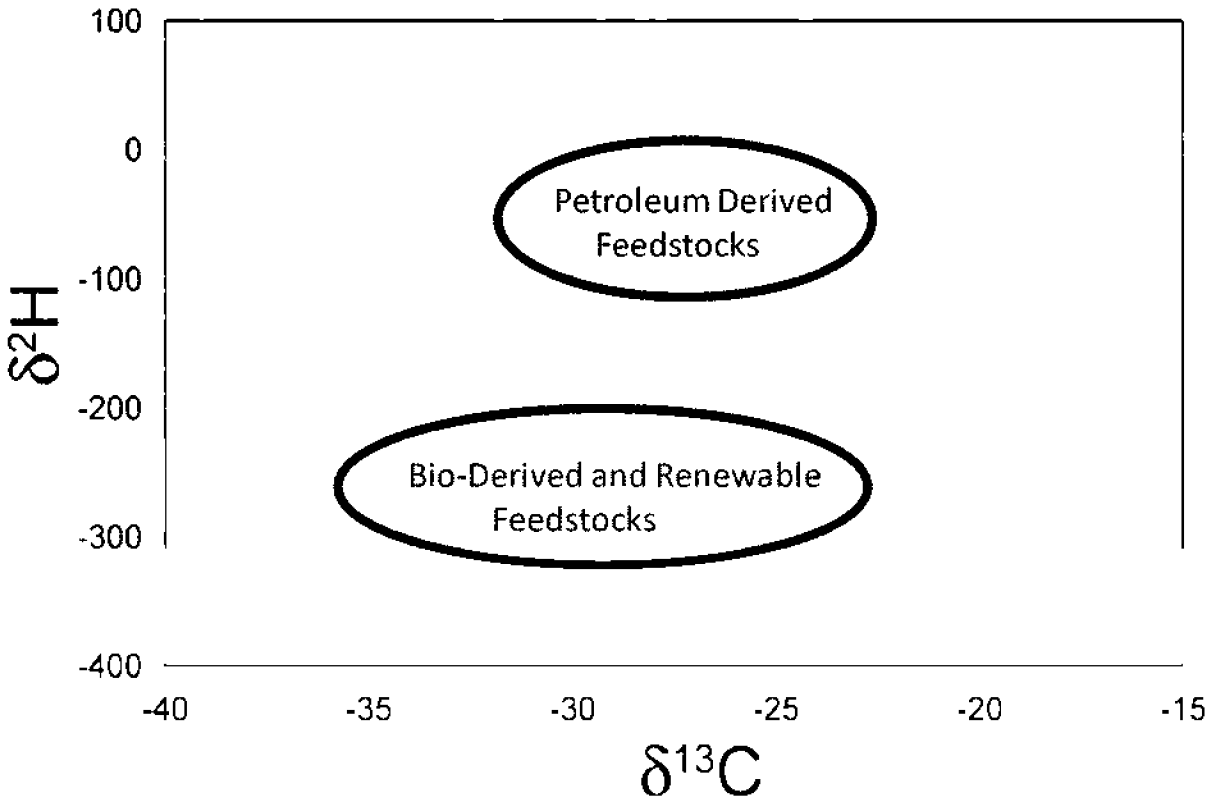

100
0
-100
-200
-300
-400
δ^2H
Petroleum Derived Feedstocks
Bio-Derived and Renewable Feedstocks
-40
-35
-30
-25
-20
-15
$\delta^{13}C$

Fig. 2

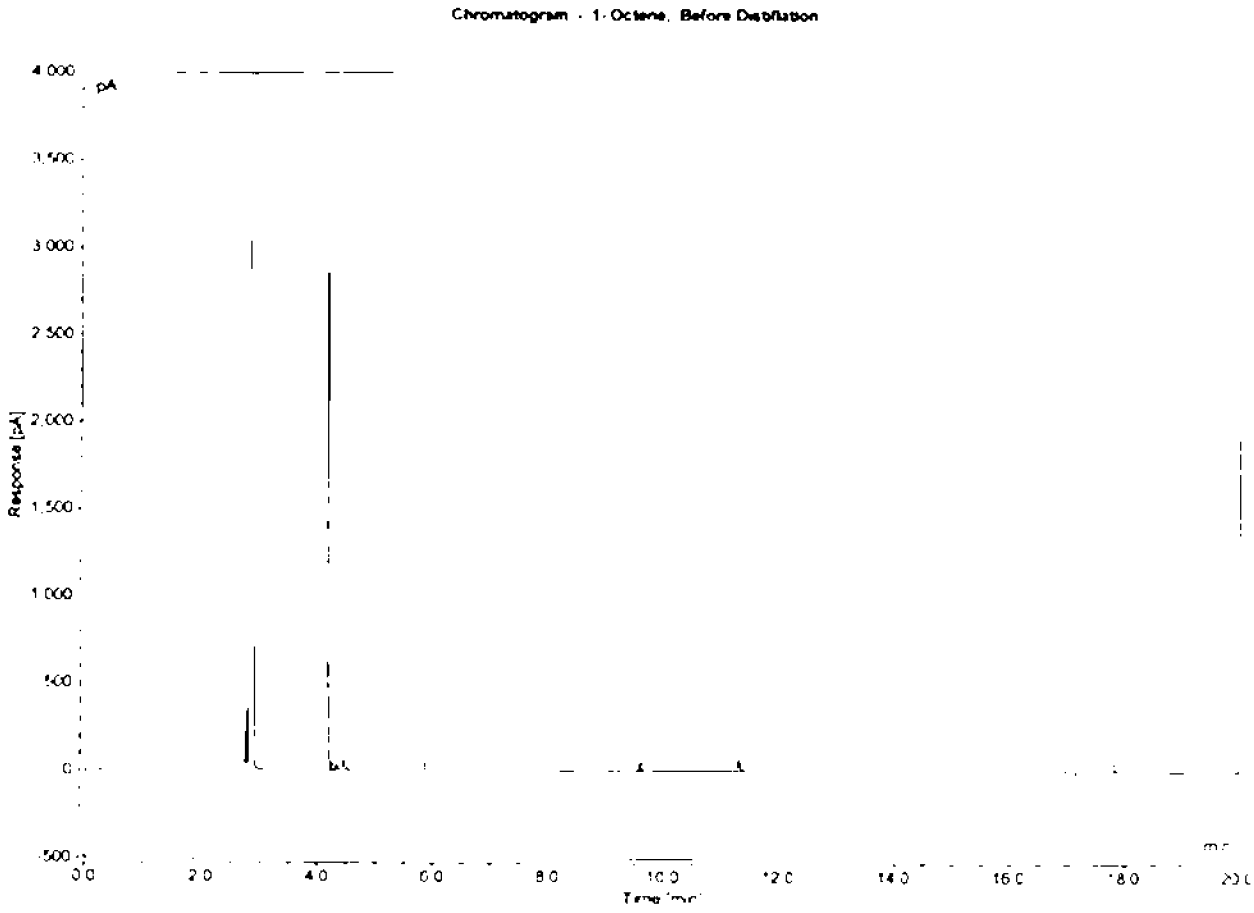

Chromatogram - 1-Octene, Before Destillation
pA
Response [pA]
4 000
3 500
3 000
2 500
2 000
1 500
1 000
500
0
-500
0.0
2.0
4.0
6.0
8.0
10.0
12.0
14.0
16.0
18.0
20.0
Time [min]

Fig. 3

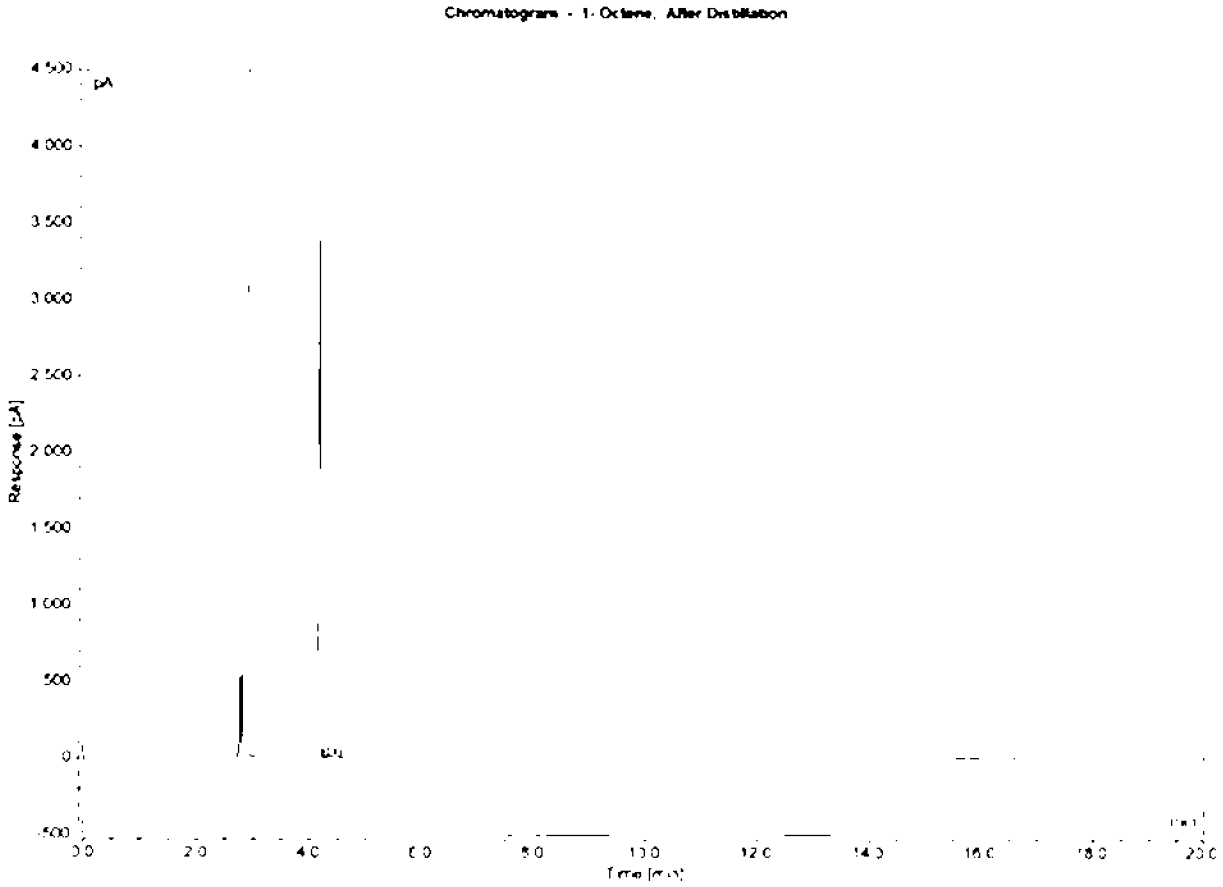

Chromatogram - 1-Octene, After Distillation
pA
4 500
4 000
3 500
3 000
2 500
2 000
1 500
1 000
500
0
-500
Response [pA]
0.0
2.0
4.0
6.0
8.0
10.0
12.0
14.0
16.0
18.0
20.0
Time [min]

Fig. 4

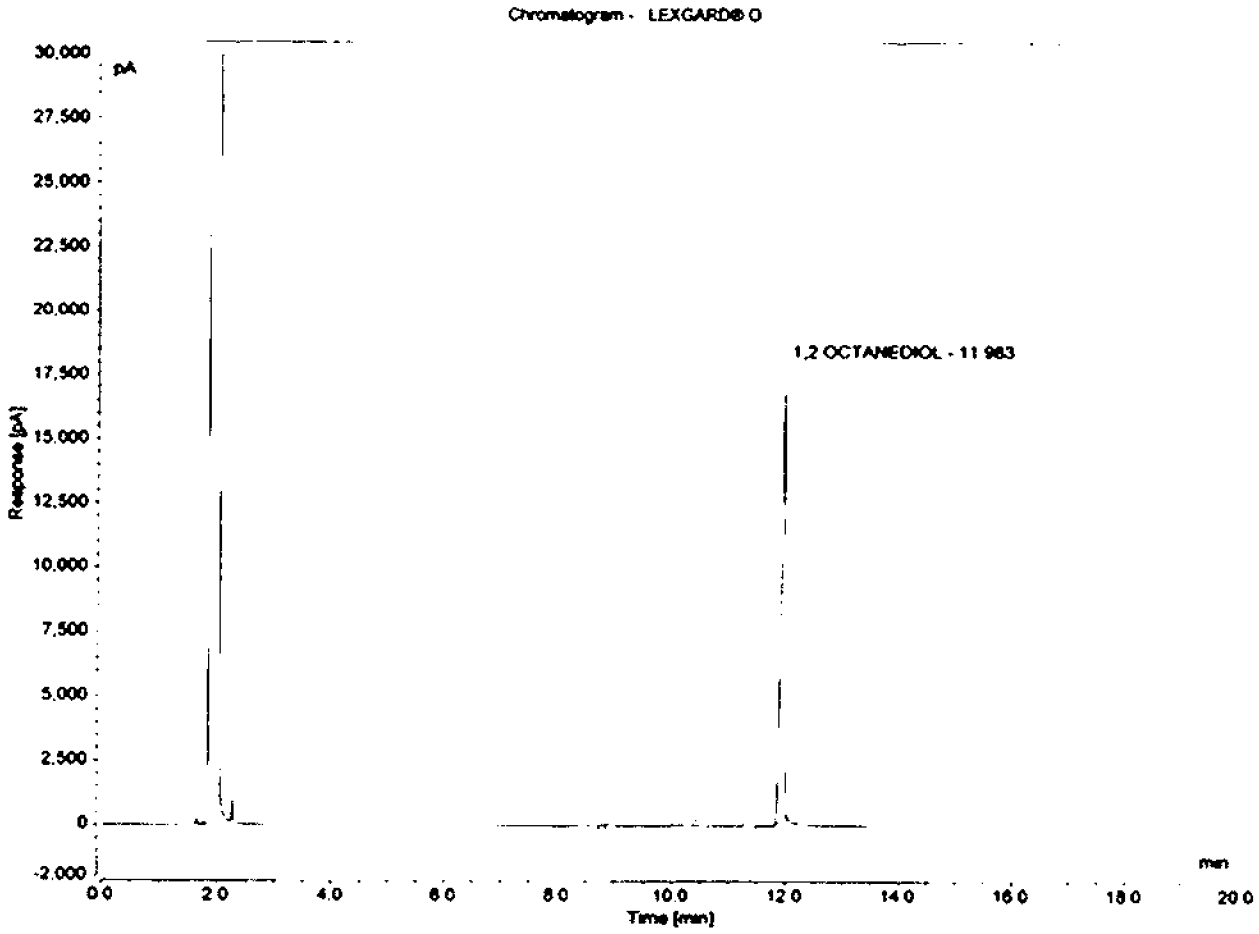

Chromatogram - LEXGARD® O
30,000
pA
27,500
25,000
22,500
20,000
17,500
15,000
12,500
10,000
7,500
5,000
2,500
0
-2,000
Response [pA]
1,2 OCTANEDIOL - 11.983
min
0.0
2.0
4.0
6.0
8.0
10.0
12.0
14.0
16.0
18.0
20.0
Time [min]

Fig. 5

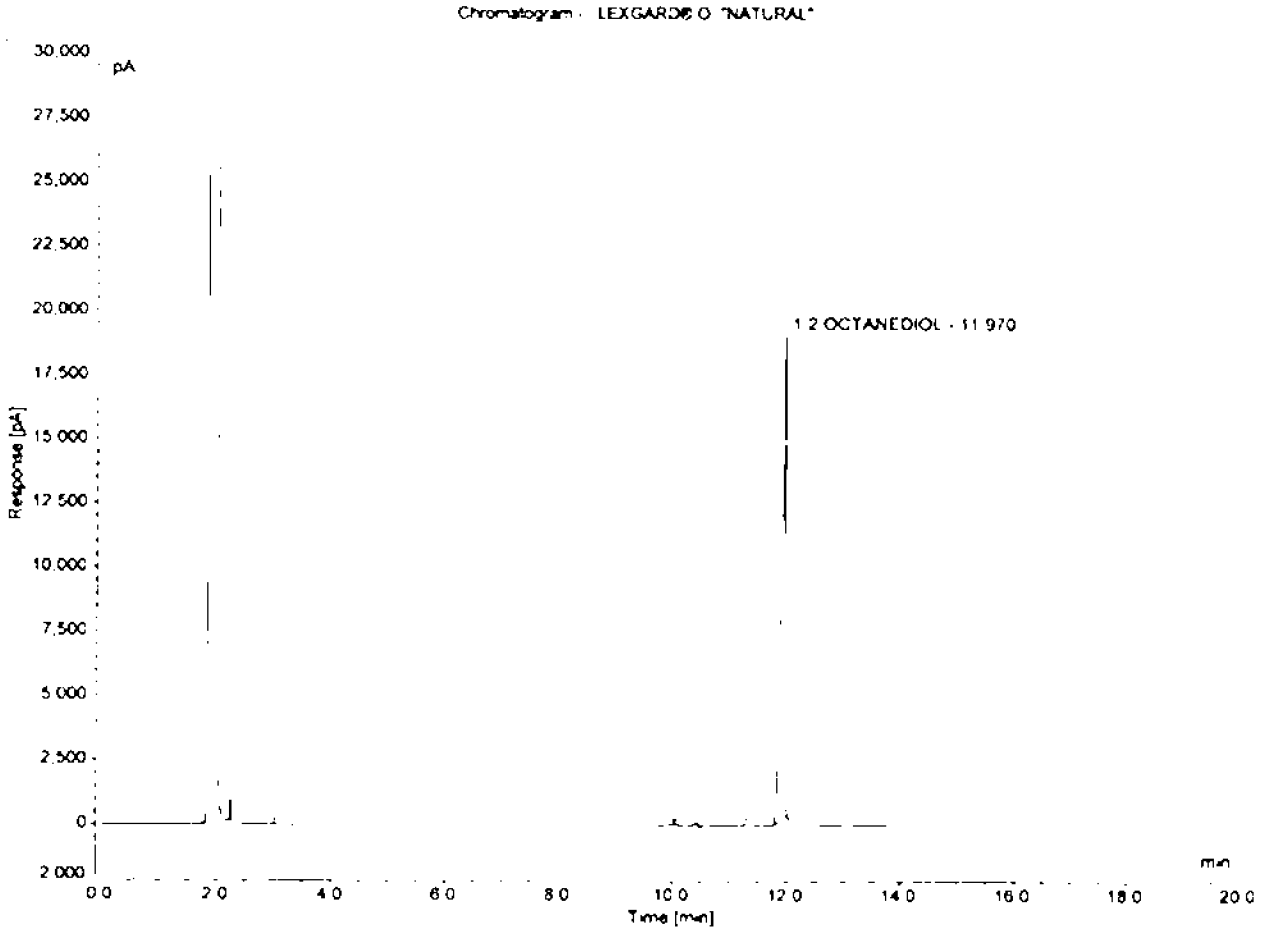

Chromatogram - LEXGARD® O "NATURAL"
30,000
pA
27,500
25,000
22,500
20,000
17,500
15,000
12,500
10,000
7,500
5,000
2,500
0
-2,000
Response [pA]
1,2-OCTANEDIOL - 11,970
min
0,0
2,0
4,0
6,0
8,0
10,0
12,0
14,0
16,0
18,0
20,0
Time [min]

Fig. 6

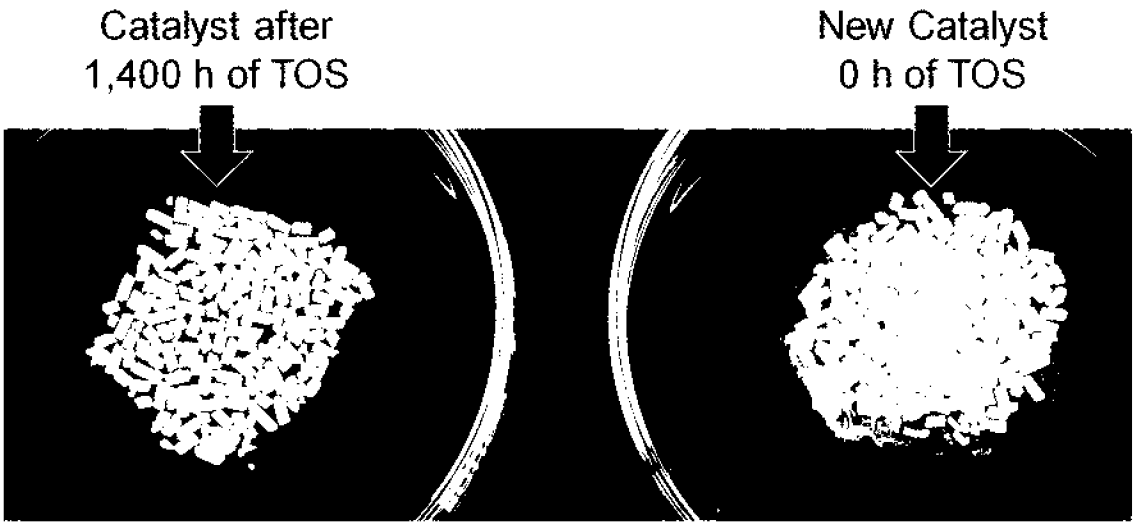


Fig. 7

DECLARATION
(Original application)

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am an original sole inventor (if only one name is listed below) or an original joint inventor (if plural names are listed below) of the subject matter that is claimed and for which a patent is sought on the invention entitled:

NATURAL 1,2-ALKANEDIOLS, COMPOSITIONS HAVING NATURAL 1,2-ALKANEDIOLS AND PROCESSES FOR MAKING THE SAME.

the specification of which is attached hereto and/or was filed on _____ as Application No. _____.

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to herein.

The above-identified application was made or was authorized to be made by me.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d), of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

FOREIGN PRIORITY APPLICATION(S)

			Priority Claimed
_____			[ ] Yes [ ] No
(Number)	(Country)	(Day/month/year filed)	
_____			[ ]Yes [ ] No
(Number)	(Country)	(Day/month/year filed)	

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional patent application(s) listed below:

PROVISIONAL PRIORITY PATENT APPLICATION(S)

		Priority Claimed
62/623,985	January 30, 2018	[X] Yes [ ] No
(Application No.)	(Filing Date)	
		[ ] Yes [ ] No
(Application No.)	(Filing Date)	

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements made in this Declaration are punishable under 18 U.S.C. §1001 by fine or imprisonment of not more than five (5) years.

Full name of first or sole inventor Rocco V. Burgo

Inventor's Signature

Date

Residence Mullica Hill, New Jersey

Citizenship United States

Post Office Address 604 Widgeon Court, Mullica Hill, New Jersey 08062

Full name of second inventor Michael E. Wright

Inventor's Signature

Date

Residence Hurricane, Utah

Citizenship United States

Post Office Address 4175 S. 1200 W., Hurricane, Utah 84737

Full name of third inventor	Gary B. Mosser
Inventor's Signature	
Date	
Residence	Tabernacle, New Jersey
Citizenship	United States
Post Office Address	5 Colonial Court, Tabernacle, New Jersey 08088

Full name of fourth inventor	Michael J. Fevola
Inventor's Signature	
Date	
Residence	Belle Mead, New Jersey
Citizenship	United States
Post Office Address	409 Township Line Road, Belle Mead, New Jersey 08502

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NUMBER	FILING or 371(c) DATE	GRP ART UNIT	FIL FEE REC'D	ATTY.DOCKET.NO	TOT CLAIMS	IND CLAIMS
16/262,902	01/30/2019		0.00	I0113-198U2	46	5

CONFIRMATION NO. 1028

FILING RECEIPT

159443
Calderone Bullock LLC
Two Greentree Centre, Suite 301
9000 Lincoln Drive East
Marlton, NJ 08053

OC000000105964857

Date Mailed: 02/19/2019

Receipt is acknowledged of this non-provisional patent application. The application will be taken up for examination in due course. Applicant will be notified as to the results of the examination. Any correspondence concerning the application must include the following identification information: the U.S. APPLICATION NUMBER, FILING DATE, NAME OF APPLICANT, and TITLE OF INVENTION. Fees transmitted by check or draft are subject to collection. Please verify the accuracy of the data presented on this receipt. **If an error is noted on this Filing Receipt, please submit a written request for a Filing Receipt Correction. Please provide a copy of this Filing Receipt with the changes noted thereon. If you received a "Notice to File Missing Parts" for this application, please submit any corrections to this Filing Receipt with your reply to the Notice. When the USPTO processes the reply to the Notice, the USPTO will generate another Filing Receipt incorporating the requested corrections**

Inventor(s)

Rocco V. Burgo, Mullica Hill, NJ;
Michael E. Wright, Hurricane, UT;
Gary B. Mosser, Tabernacle, NJ;
Michael J. Fevola, Belle Mead, NJ;

Applicant(s)

Inolex Investment Corporation, Wilmington, DE

Assignment For Published Patent Application

Inolex Investment Corporation

Power of Attorney: None

Domestic Priority data as claimed by applicant

This appln claims benefit of 62/623,985 01/30/2018

Foreign Applications for which priority is claimed (You may be eligible to benefit from the **Patent Prosecution Highway** program at the USPTO. Please see http://www.uspto.gov for more information.) - None.
Foreign application information must be provided in an Application Data Sheet in order to constitute a claim to foreign priority. See 37 CFR 1.55 and 1.76.

Permission to Access Application via Priority Document Exchange: Yes

Permission to Access Search Results: Yes

Applicant may provide or rescind an authorization for access using Form PTO/SB/39 or Form PTO/SB/69 as appropriate.

If Required, Foreign Filing License Granted: 02/15/2019

The country code and number of your priority application, to be used for filing abroad under the Paris Convention, is **US 16/262,902**

Projected Publication Date: To Be Determined - pending completion of Missing Parts

Non-Publication Request: No

Early Publication Request: No

Title

Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same

Preliminary Class

Statement under 37 CFR 1.55 or 1.78 for AIA (First Inventor to File) Transition Applications: No

PROTECTING YOUR INVENTION OUTSIDE THE UNITED STATES

Since the rights granted by a U.S. patent extend only throughout the territory of the United States and have no effect in a foreign country, an inventor who wishes patent protection in another country must apply for a patent in a specific country or in regional patent offices. Applicants may wish to consider the filing of an international application under the Patent Cooperation Treaty (PCT). An international (PCT) application generally has the same effect as a regular national patent application in each PCT-member country. The PCT process **simplifies** the filing of patent applications on the same invention in member countries, but **does not result** in a grant of "an international patent" and does not eliminate the need of applicants to file additional documents and fees in countries where patent protection is desired.

Almost every country has its own patent law, and a person desiring a patent in a particular country must make an application for patent in that country in accordance with its particular laws. Since the laws of many countries differ in various respects from the patent law of the United States, applicants are advised to seek guidance from specific foreign countries to ensure that patent rights are not lost prematurely.

Applicants also are advised that in the case of inventions made in the United States, the Director of the USPTO must issue a license before applicants can apply for a patent in a foreign country. The filing of a U.S. patent application serves as a request for a foreign filing license. The application's filing receipt contains further information and guidance as to the status of applicant's license for foreign filing.

Applicants may wish to consult the USPTO booklet, "General Information Concerning Patents" (specifically, the section entitled "Treaties and Foreign Patents") for more information on timeframes and deadlines for filing foreign patent applications. The guide is available either by contacting the USPTO Contact Center at 800-786-9199, or it can be viewed on the USPTO website at http://www.uspto.gov/web/offices/pac/doc/general/index.html.

For information on preventing theft of your intellectual property (patents, trademarks and copyrights), you may wish to consult the U.S. Government website, http://www.stopfakes.gov. Part of a Department of Commerce initiative, this website includes self-help "toolkits" giving innovators guidance on how to protect intellectual property in specific countries such as China, Korea and Mexico. For questions regarding patent enforcement issues, applicants may call the U.S. Government hotline at 1-866-999-HALT (1-866-999-4258).

LICENSE FOR FOREIGN FILING UNDER

Title 35, United States Code, Section 184

Title 37, Code of Federal Regulations, 5.11 & 5.15

GRANTED

The applicant has been granted a license under 35 U.S.C. 184, if the phrase "IF REQUIRED, FOREIGN FILING LICENSE GRANTED" followed by a date appears on this form. Such licenses are issued in all applications where the conditions for issuance of a license have been met, regardless of whether or not a license may be required as set forth in 37 CFR 5.15. The scope and limitations of this license are set forth in 37 CFR 5.15(a) unless an earlier license has been issued under 37 CFR 5.15(b). The license is subject to revocation upon written notification. The date indicated is the effective date of the license, unless an earlier license of similar scope has been granted under 37 CFR 5.13 or 5.14.

This license is to be retained by the licensee and may be used at any time on or after the effective date thereof unless it is revoked. This license is automatically transferred to any related applications(s) filed under 37 CFR 1.53(d). This license is not retroactive.

The grant of a license does not in any way lessen the responsibility of a licensee for the security of the subject matter as imposed by any Government contract or the provisions of existing laws relating to espionage and the national security or the export of technical data. Licensees should apprise themselves of current regulations especially with respect to certain countries, of other agencies, particularly the Office of Defense Trade Controls, Department of State (with respect to Arms, Munitions and Implements of War (22 CFR 121-128)); the Bureau of Industry and Security, Department of Commerce (15 CFR parts 730-774); the Office of Foreign AssetsControl, Department of Treasury (31 CFR Parts 500+) and the Department of Energy.

NOT GRANTED

No license under 35 U.S.C. 184 has been granted at this time, if the phrase "IF REQUIRED, FOREIGN FILING LICENSE GRANTED" DOES NOT appear on this form. Applicant may still petition for a license under 37 CFR 5.12, if a license is desired before the expiration of 6 months from the filing date of the application. If 6 months has lapsed from the filing date of this application and the licensee has not received any indication of a secrecy order under 35 U.S.C. 181, the licensee may foreign file the application pursuant to 37 CFR 5.15(b).

SelectUSA

The United States represents the largest, most dynamic marketplace in the world and is an unparalleled location for business investment, innovation, and commercialization of new technologies. The U.S. offers tremendous resources and advantages for those who invest and manufacture goods here. Through SelectUSA, our nation works to promote and facilitate business investment. SelectUSA provides information assistance to the international investor community; serves as an ombudsman for existing and potential investors; advocates on behalf of U.S. cities, states, and regions competing for global investment; and counsels U.S. economic development organizations on investment attraction best practices. To learn more about why the United States is the best country in the world to develop technology, manufacture products, deliver services, and grow your business, visit http://www.SelectUSA.gov or call +1-202-482-6800.

PATENT APPLICATION FEE DETERMINATION RECORD

Substitute for Form PTO-875

Application or Docket Number: 16/262,902

APPLICATION AS FILED - PART I

FOR	(Column 1) NUMBER FILED	(Column 2) NUMBER EXTRA	SMALL ENTITY RATE($)	SMALL ENTITY FEE($)	OR	OTHER THAN SMALL ENTITY RATE($)	OTHER THAN SMALL ENTITY FEE($)
BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A	N/A			N/A	300
SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A	N/A			N/A	660
EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A	N/A			N/A	760
TOTAL CLAIMS (37 CFR 1.16(i))	46 minus 20 =	* 26			OR	x 100 =	2600
INDEPENDENT CLAIMS (37 CFR 1.16(h))	5 minus 3 =	* 2				x 460 =	920
APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $310 ($155 for small entity) for each additional 50 sheets or fraction thereof. See 35 U.S.C. 41(a)(1)(G) and 37 CFR 1.16(s).						0.00
MULTIPLE DEPENDENT CLAIM PRESENT (37 CFR 1.16(j))							0.00
* If the difference in column 1 is less than zero, enter "0" in column 2.			TOTAL			TOTAL	5240

APPLICATION AS AMENDED - PART II

		(Column 1) CLAIMS REMAINING AFTER AMENDMENT		(Column 2) HIGHEST NUMBER PREVIOUSLY PAID FOR	(Column 3) PRESENT EXTRA	SMALL ENTITY RATE($)	SMALL ENTITY ADDITIONAL FEE($)	OR	OTHER THAN SMALL ENTITY RATE($)	OTHER THAN SMALL ENTITY ADDITIONAL FEE($)
AMENDMENT A	Total (37 CFR 1.16(i))	*	Minus	**	=	x =		OR	x =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=	x =		OR	x =	
	Application Size Fee (37 CFR 1.16(s))									
	FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							OR		
						TOTAL ADD'L FEE		OR	TOTAL ADD'L FEE	

		(Column 1) CLAIMS REMAINING AFTER AMENDMENT		(Column 2) HIGHEST NUMBER PREVIOUSLY PAID FOR	(Column 3) PRESENT EXTRA	SMALL ENTITY RATE($)	SMALL ENTITY ADDITIONAL FEE($)	OR	OTHER THAN SMALL ENTITY RATE($)	OTHER THAN SMALL ENTITY ADDITIONAL FEE($)
AMENDMENT B	Total (37 CFR 1.16(i))	*	Minus	**	=	x =		OR	x =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=	x =		OR	x =	
	Application Size Fee (37 CFR 1.16(s))									
	FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							OR		
						TOTAL ADD'L FEE		OR	TOTAL ADD'L FEE	

* If the entry in column 1 is less than the entry in column 2, write "0" in column 3.

** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 20, enter "20".

*** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 3, enter "3".

The "Highest Number Previously Paid For" (Total or Independent) is the highest found in the appropriate box in column 1.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NUMBER	FILING OR 371(C) DATE	FIRST NAMED APPLICANT	ATTY. DOCKET NO./TITLE
16/262,902	01/30/2019	Rocco V. Burgo	I0113-198U2

CONFIRMATION NO. 1028

159443
Calderone Bullock LLC
Two Greentree Centre, Suite 301
9000 Lincoln Drive East
Marlton, NJ 08053

FORMALITIES LETTER

OC000000105964858

Date Mailed: 02/19/2019

NOTICE TO FILE MISSING PARTS OF NONPROVISIONAL APPLICATION

FILED UNDER 37 CFR 1.53(b)

Filing Date Granted

Items Required To Avoid Abandonment:

An application number and filing date have been accorded to this application. The item(s) indicated below, however, are missing. Applicant is given **TWO MONTHS** from the date of this Notice within which to file all required items below to avoid abandonment. Extensions of time may be obtained by filing a petition accompanied by the extension fee under the provisions of 37 CFR 1.136(a).

- The statutory basic filing fee is missing.
- The application search fee must be submitted.
- The application examination fee must be submitted.

The application is informal since it does not comply with the regulations for the reason(s) indicated below.

The required item(s) identified below must be timely submitted to avoid abandonment:

- Replacement drawings in compliance with 37 CFR 1.84 and 37 CFR 1.121(d) are required. The drawings submitted are not acceptable because:
  - The drawings submitted to the Office are not electronically reproducible because portions of figures 3-6 are missing and/or blurry.

Applicant is cautioned that correction of the above items may cause the specification and drawings page count to exceed 100 pages. If the specification and drawings exceed 100 pages, applicant will need to submit the required application size fee.

- Additional claim fees of $ **3520** as an undiscounted entity, including any required multiple dependent claim fee, are required. Applicant must submit the additional claim fees or cancel the additional claims for which fees are due.
- Surcharge as set forth in 37 CFR 1.16(f) must be submitted.
  The surcharge is due for any one of:
  - late submission of the basic filing fee, search fee, or examination fee,
  - late submission of inventor's oath or declaration,
  - filing an application that does not contain at least one claim on filing, or
  - submission of an application filed by reference to a previously filed application.

SUMMARY OF FEES DUE:

The fee(s) required within **TWO MONTHS** from the date of this Notice to avoid abandonment is/are itemized below. No entity status discount is in effect. If applicant is qualified for small entity status, a written assertion of small entity status must be submitted to establish small entity status. (See 37 CFR 1.27). If applicant is qualified for micro entity status, an acceptable Certification of Micro Entity Status must be submitted to establish micro entity status. (See 37 CFR 1.29 and forms PTO/SB/15A and 15B.)

- $ **300** basic filing fee.
- $ **160** surcharge.
- $ **660** search fee.
- $ **760** examination fee.
- $ **920** for **2** independent claims over 3.
- $ **2600** for **26** total claims over 20.
- $(**0**) previous unapplied payment amount.
- $ **5400** TOTAL FEE BALANCE DUE.

Items Required To Avoid Processing Delays:

Applicant is notified that the above-identified application contains the deficiencies noted below. No period for reply is set forth in this notice for correction of these deficiencies. However, if a deficiency relates to the inventor's oath or declaration, the applicant must file an oath or declaration in compliance with 37 CFR 1.63, or a substitute statement in compliance with 37 CFR 1.64, executed by or with respect to each actual inventor no later than the expiration of the time period set in the "Notice of Allowability" to avoid abandonment. See 37 CFR 1.53(f).

- A properly executed inventor's oath or declaration has not been received for the following inventor(s):
  Rocco V. Burgo
  Michael E. Wright
  Gary B. Mosser
  Michael J. Fevola

Replies must be received in the USPTO within the set time period or must include a proper Certificate of Mailing or Transmission under 37 CFR 1.8 with a mailing or transmission date within the set time period. For more information and a suggested format, see Form PTO/SB/92 and MPEP 512.

Replies should be mailed to:

Mail Stop Missing Parts
Commissioner for Patents
P.O. Box 1450
Alexandria VA 22313-1450

Registered users of EFS-Web may alternatively submit their reply to this notice via EFS-Web, including a copy of this Notice and selecting the document description "Applicant response to Pre-Exam Formalities Notice". https://sportal.uspto.gov/authenticate/AuthenticateUserLocalEPF.html

For more information about EFS-Web please call the USPTO Electronic Business Center at 1-866-217-9197 or visit our website at http://www.uspto.gov/ebc.

If you are not using EFS-Web to submit your reply, you must include a copy of this notice.

Questions about the contents of this notice and the requirements it sets forth should be directed to the Office of Data Management, Application Assistance Unit, at **(571) 272-4000** or **(571) 272-4200** or **1-888-786-0101**.

/fhadera/

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Non-Provisional Patent Application of: Inolex Investment Corporation			
Conf. No.:	1028	Group Art Unit:	Not Yet Assigned
Appln. No.:	16/262,902	Examiner:	
Filing Date:	January 30, 2019	Attorney Docket No.:	I0113-198U2
Title:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making The Same		

AUTHORIZATION OF PAYMENT OF ALL EXTENSION OF TIME FEES

In the event that a Petition for Extension of Time is required at any time during the prosecution of the above-captioned application, but not submitted, the Commissioner is hereby authorized to charge any extension fee under 37 C.F.R. § 1.136(a) to Deposit Account No. 60-2689 (Billing No. I0113.0224).

Respectfully submitted,

Inolex Investment Corporation

8 March 2019 (Date) By: 

KRISTYNE A. BULLOCK
Registration No. 42,371
CALDERONE BULLOCK LLC
Two Greentree Centre
9000 Lincoln Drive East, Suite 301
Marlton, NJ 08053
Telephone: (856) 446-6047
Direct Dial: (856) 446-6043
Facsimile: (856) 267-5370
E-Mail: kbullock@calderonebullock.com

KAB/jxv

Electronic Acknowledgement Receipt

EFS ID:	35369821
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	159443
Filer:	Kristyne Alison Bullock
Filer Authorized By:	
Attorney Docket Number:	I0113-198U2
Receipt Date:	08-MAR-2019
Filing Date:	30-JAN-2019
Time Stamp:	18:35:19
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Authorization for Extension of Time all replies	198U2.pdf	44442 b046cc3fc05f77ec30aefbaf43229b8676c43ebb	no	1

Warnings:

Information:	
Total Files Size (in bytes):	44442

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

ELECTRONICALLY FILED

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Patent Application of: Inolex Investment Corporation			
Conf. No.:	1028	Group Art Unit:	Not Yet Assigned
Appln. No.:	16/262,902	Examiner:	
Filing Date:	January 30, 2019	Attorney Docket No.:	I0113-198U2

Title: Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same

PETITION FOR EXTENSION OF TIME UNDER (37 C.F.R. § 1.136(a))

Applicant in the above-identified application hereby petitions for a two-month extension of time to and including June 19, 2019 for responding to the Notice to File Missing Parts of February 19, 2019. A Response to Missing Parts is being filed simultaneously with the filing of this Petition.

[X] Applicant herewith charges the extension fee in the amount of $600 to credit account Calderone Bullock American Express (Billing No. I0113.0251).

[X] The Commissioner is hereby authorized to charge any deficiencies and to credit any overpayments to Deposit Account No. 60-2689 (Billing No. I0113.0251).

Respectfully submitted,

INOLEX INVESTMENT CORPORATION

6/19/19 By: _____
(Date)

LYNDA L. CALDERONE
Registration No. 35,837
CALDERONE BULLOCK LLC
Two Greentree Centre
9000 Lincoln Drive East, Suite 301
Marlton, NJ 08053
Telephone: (856) 446-6047
Direct Dial: (856) 446-6045
Facsimile: (856) 267-5370
E-Mail: lcalderone@calderonebullock.com

LLC:llc

Electronic Patent Application Fee Transmittal

Application Number:	16262902
Filing Date:	30-Jan-2019
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Filer:	lynda Leigh Calderone
Attorney Docket Number:	I0113-198U2

Filed as Large Entity

Filing Fees for Utility under 35 USC 111(a)

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
UTILITY APPLICATION FILING	1011	1	300	300
UTILITY SEARCH FEE	1111	1	660	660
UTILITY EXAMINATION FEE	1311	1	760	760
Pages:				
Claims:				
CLAIMS IN EXCESS OF 20	1202	3	100	300
Miscellaneous-Filing:				
LATE FILING FEE FOR OATH OR DECLARATION	1051	1	160	160

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
Extension-of-Time:				
Extension - 2 months with $0 paid	1252	1	600	600
Miscellaneous:				
		Total in USD ($)		**2780**

Electronic Acknowledgement Receipt

EFS ID:	36351215
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	159443
Filer:	lynda Leigh Calderone
Filer Authorized By:	
Attorney Docket Number:	I0113-198U2
Receipt Date:	19-JUN-2019
Filing Date:	30-JAN-2019
Time Stamp:	17:35:47
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	CARD
Payment was successfully received in RAM	$2780
RAM confirmation Number	062019INTEFSW17364600
Deposit Account	602689
Authorized User	Lynda Calderone

The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows:

37 CFR 1.17 (Patent application and reexamination processing fees)

37 CFR 1.19 (Document supply fees)

37 CFR 1.20 (Post Issuance fees)

37 CFR 1.21 (Miscellaneous fees and charges)

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Miscellaneous Incoming Letter	responsetomissingparts.pdf	177694 6898f23fc7891f384650a29ce812fc4c89f82d3e	no	4
Warnings:					
Information:					
2	Drawings-only black and white line drawings	Replacementdrawingsp.pdf	875093 110c148ef38f40d010d564790a55e5b2cc14ea92	no	7
Warnings:					
Information:					
3	Preliminary Amendment	secondprelamend.pdf	191167 b391b6d26c20199a800d62fae0b25634953c22ea	no	5
Warnings:					
Information:					
4	Extension of Time	Extensionoftime.pdf	50307 2bb969a52fdb9362216af7b468e3de3fbce607e9	no	1
Warnings:					
Information:					
5	Fee Worksheet (SB06)	fee-info.pdf	40845 b50299bc268db036fad42d0fd076b54857bdf3f9	no	2
Warnings:					
Information:					
		Total Files Size (in bytes):	1335106		

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

ELECTRONICALLY FILED

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Patent Application of: Inolex Investment Corporation			
Conf. No.:	1028	Group Art Unit:	Not Yet Assigned
Appln. No.:	16/262,902	Examiner:	
Filing Date:	January 30, 2019	Attorney Docket No.:	I0113-198U2
Title:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same		

TRANSMITTAL OF MISSING PARTS - FILING DATE GRANTED

This is in response to the Notice to File Missing Parts dated February 19, 2019. This response is being timely filed on or before June 19, 2018 in view of the simultaneous filing of a Petition for Extension of Time for two months, including authorization to pay the appropriate fee, we submit the following:

[ ] Copy of the Notice to File Missing Parts -- Filing Date Granted, mailed _____;

[ ] [ ] Inventor(s) Declaration; [ ] Inventor(s) Declaration and Power of Attorney;
[ ] Inventor(s) Oath; or [ ] Substitute Statement.

[ ] Appointment of Attorney by Applicant/Assignee.

[ ] General Power of Attorney and 3.73(c) Statement.

[ ] Revocation and Appointment of Attorney and Change of Correspondence Address.

[X] [ ] Formal Drawings: ____ Sheets; Replacement Drawings: 7 Sheets.

[ ] Substitute Application Data Sheet.

[ ] Verified English translation of application as filed.

[ ] Priority document(s): Certified copy(ies) of __ patent application No(s). __, filed in __ on __ from which priority is claimed under 35 U.S.C. § 119.

[ ] Copies of Executed [ ] Inventor Declaration from prior Application No. _____; [ ] Declaration and Power of Attorney from Prior Application No. _____.

[ ] Formal drawings: __ sheets

[ ] Information Disclosure Statement, PTO/SB/08A or B and cited references.

[ ] Nucleotide and/or Amino Acid Sequence Submission, including:

[ ] Computer readable copy [ ] Paper Copy [ ] Verified Statement

[ ] Microfiche computer program (Appendix).

[X] Petition for Extension of Time along with requisite fee.

[ ] Request for Refund.

[ ] Request for Corrected Filing Receipt. Attached is a marked-up copy of the Filing Receipt showing corrections.

[ ] Applicant(s), by its/their undersigned attorney, claim(s) Small Entity Status under 37 C.F.R. §1.27 as [ ] an Independent Inventor, or [ ] a Small Business Concern, or [ ] a Non-Profit Organization.

[ ] Micro Entity Status [ ] is hereby claimed by Applicant(s) and a Certification of Micro Entity Status is enclosed; [ ] has previously been claimed/established.

[X] Utility Non-Provisional Patent Application Filing Fees. The application filing fees were not paid at filing and are being paid herewith and are calculated as follows:

			MICRO ENTITY			SMALL ENTITY			LARGE ENTITY	
CLAIMS	NO. FILED	NO. EXTRA	BASIC FILING, SEARCH, AND EXAMINATION FEES:	$430		BASIC FILING, SEARCH, AND EXAMINATION FEES:	$ 785		BASIC FILING, SEARCH, AND EXAMINATION FEES:	$1,720
Total	23-20 =	3	X $ 25	$	OR	X $ 50	$	OR	X $100	$ 300
Independent	2 - 3=	0	X $115	$	OR	X $230	$	OR	X $460	$ 0
[0] Multiple Dependent Claims			X $205	$	OR	X $410	$	OR	X $820	$ 0
[ ] Application Size Fee (if specification and drawings exceed 100 sheets)										
Total sheets	-100	Add'l sheets	# of sets of 50 (round up to whole number) /50 =	X $100 =	$	OR	X $200 =	$	OR	X $400	$
Late Fee For Filing Fees, Inventor Oath or Declaration			$ 40	$	OR	$ 80	$	OR	$160	$ 160
Late Fee For Non-English Specification			$ 35	$	OR	$ 70	$	OR	$140	$ 0
			TOTAL	$	OR	TOTAL	$	OR	TOTAL	$2190

[X] Preliminary Amendment.

[ ] The application filing fees were paid upon filing and additional claim fees resulting from the Preliminary Amendment are calculated as follows:

					MICRO ENTITY		SMALL ENTITY		LARGE ENTITY	
	NO. OF CLAIMS AFTER AMEND.		HIGHEST NO. PREVIOUSLY PAID FOR	EXTRA	RATE	ADDIT. FEE	RATE	ADDIT. FEE	RATE	ADDIT. FEE
TOTAL		-			X $25	$	X $50	$	X $100	
INDEP.		-			X $115	$	X$230	$	X $460	
[ ] 1st PRESENTATION OF MULTIPLE DEPENDENT CLAIMS					$205		$410		$820	
					TOTAL	$	TOTAL	$	TOTAL	

[ ] Other:

[X] Applicant(s) herewith charge the following fees in the total amount of $2790 as calculated above to the credit card account, Calderone Bullock American Express (Billing No. I0113.0251):

[X] Surcharge of $160 for late filing of filing fee, search fee, examination fee or oath or declaration.

[ ] Surcharge of $_____ for late filing of English Specification.

[X] Extension of Time fee in the amount of $600 (Petition for Extension of Time is attached).

[X] Application Filing Fees in the amount of $2190 calculated above.

[ ] Additional Claims Fees resulting from Preliminary Amendment.

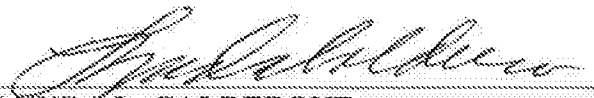
[X] The Commissioner is hereby authorized to charge and/or credit Deposit Account No. 60-2689 (Billing No. I0113.0251) as noted below.

[X] Any overpayments or deficiencies in the above-calculated fee(s).

[X] Any additional fees required under 37 C.F.R. § 1.16 or § 1.17.

[X] In the event that a Petition for Extension of Time is required during the prosecution of this application, but not submitted, please charge any extension fee under 37 C.F.R. § 1.136(a) to our Deposit Account noted above.

CORRESPONDENCE ADDRESS

6/19/19 By: 

(Date)

LYNDA L. CALDERONE
Registration No. 35,837
CALDERONE BULLOCK LLC
Two Greentree Centre
9000 Lincoln Drive East, Suite 301
Marlton, NJ 08053
Telephone: (856) 446-6047
Direct Dial: (856) 446-6045
Facsimile: (856) 267-5370
E-Mail: lcalderone@calderonebullock.com

[X] Customer Number or Bar Code Label: **159443**

LLC:llc
Enclosures

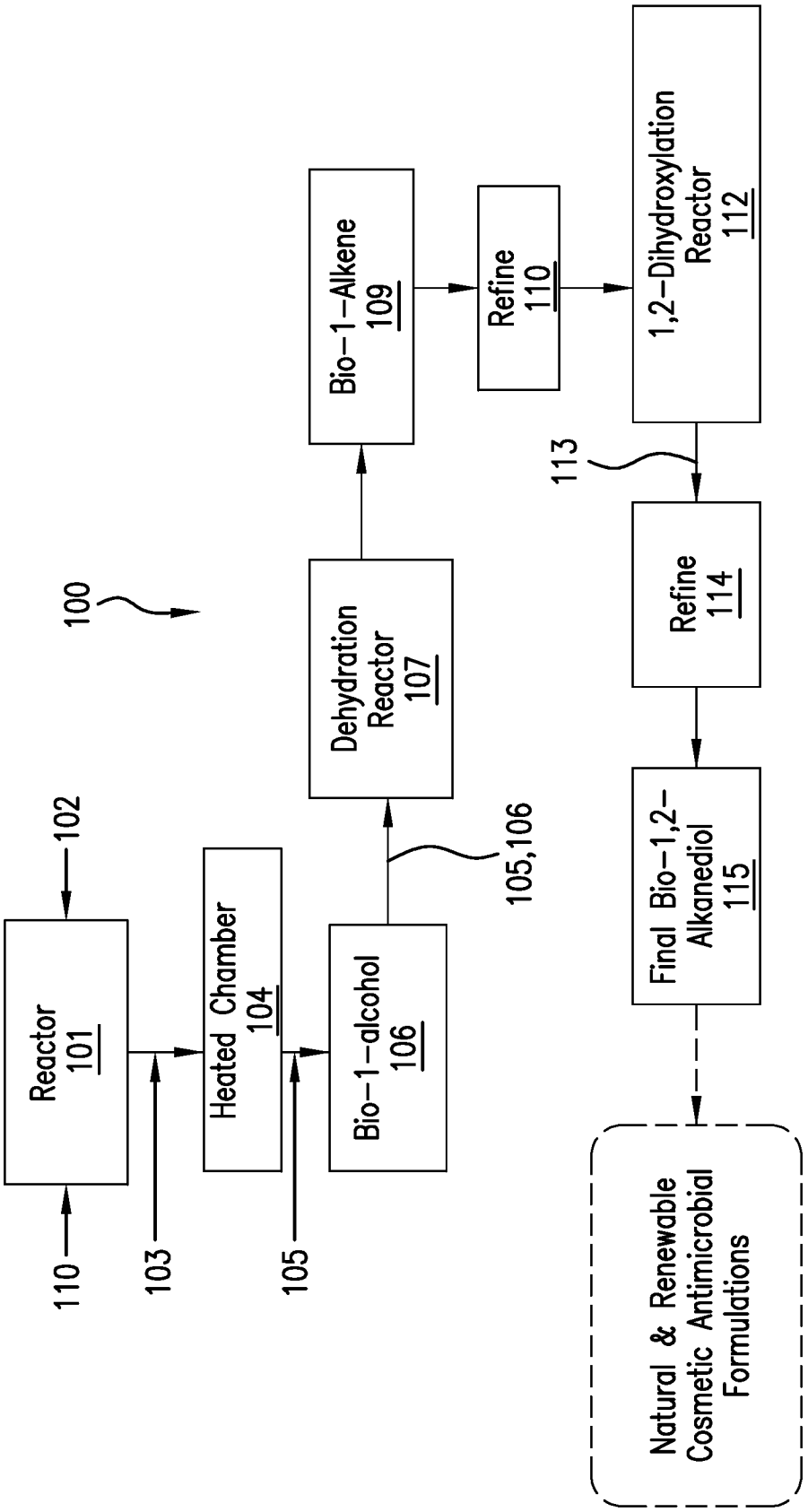

100
Reactor 101
102
110
Heated Chamber 104
103
Bio–1–alcohol 106
105
105,106
Dehydration Reactor 107
Bio–1–Alkene 109
Refine 110
1,2–Dihydroxylation Reactor 112
113
Refine 114
Final Bio–1,2–Alkanediol 115
Natural & Renewable Cosmetic Antimicrobial Formulations

FIG. 1

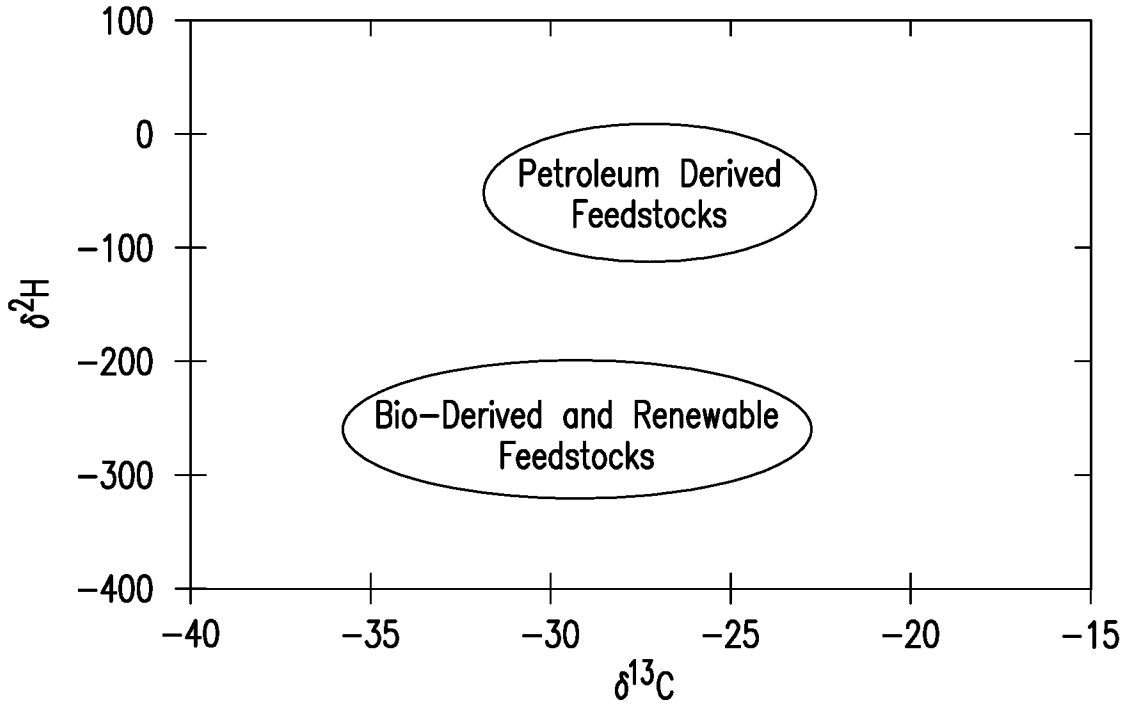

100
0
−100
−200
−300
−400
δ^2H
−40
−35
−30
−25
−20
−15
$\delta^{13}C$
Petroleum Derived Feedstocks
Bio−Derived and Renewable Feedstocks

FIG.2

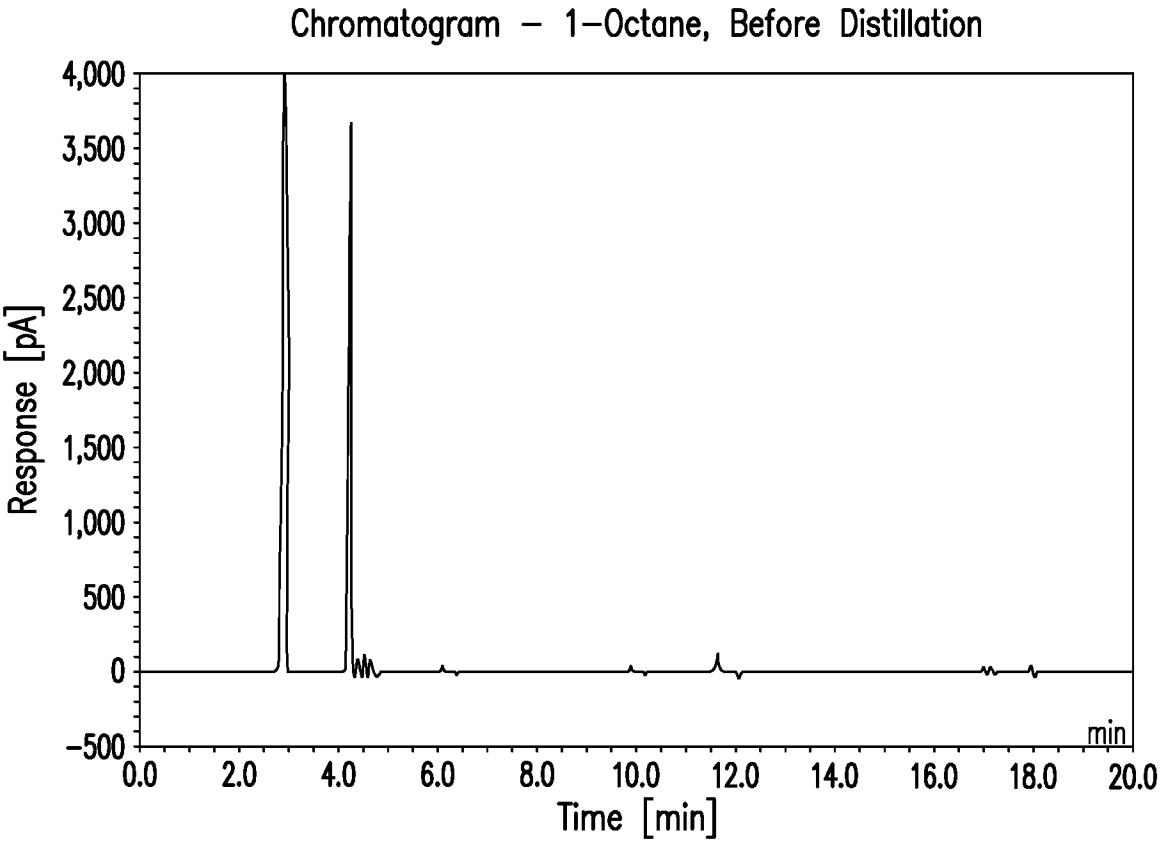

Chromatogram – 1–Octane, Before Distillation
Response [pA]
4,000
3,500
3,000
2,500
2,000
1,500
1,000
500
0
-500
min
0.0
2.0
4.0
6.0
8.0
10.0
12.0
14.0
16.0
18.0
20.0
Time [min]

FIG.3

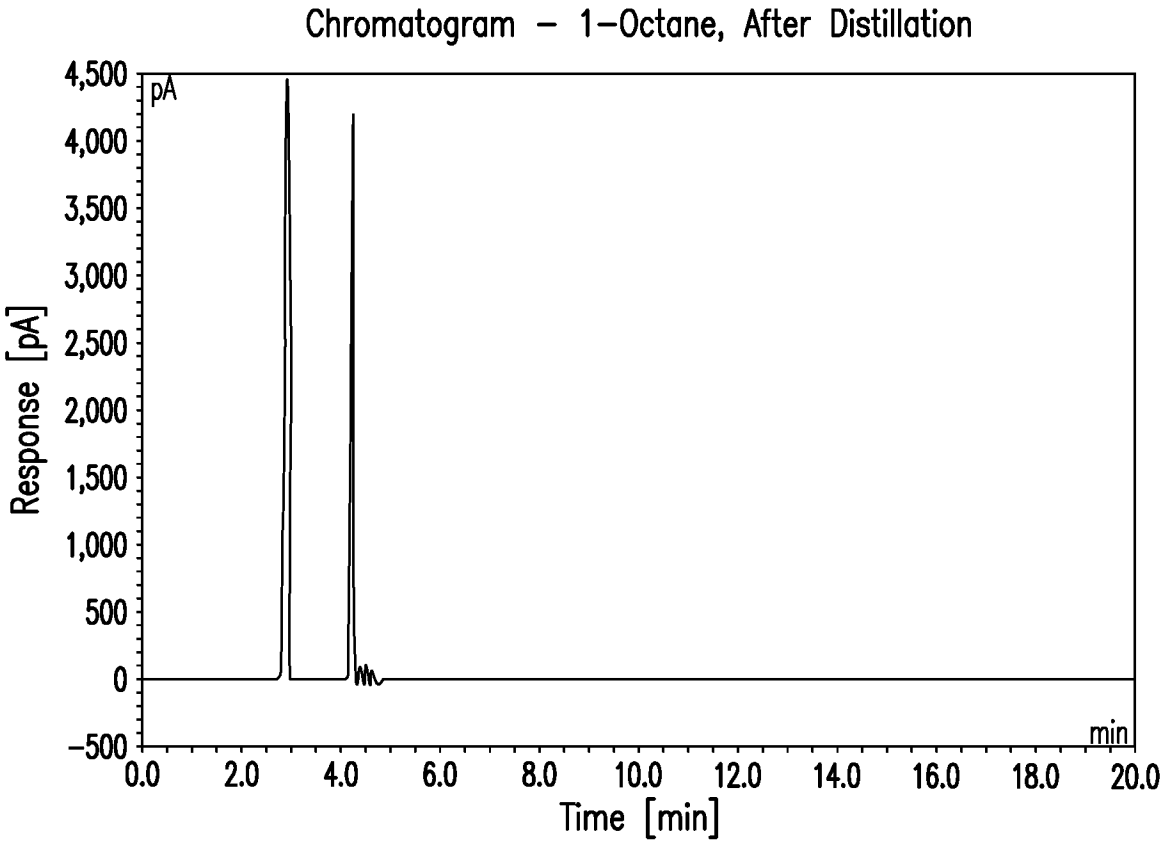

Chromatogram – 1–Octane, After Distillation
pA
Response [pA]
4,500
4,000
3,500
3,000
2,500
2,000
1,500
1,000
500
0
–500
min
0.0
2.0
4.0
6.0
8.0
10.0
12.0
14.0
16.0
18.0
20.0
Time [min]

FIG.4

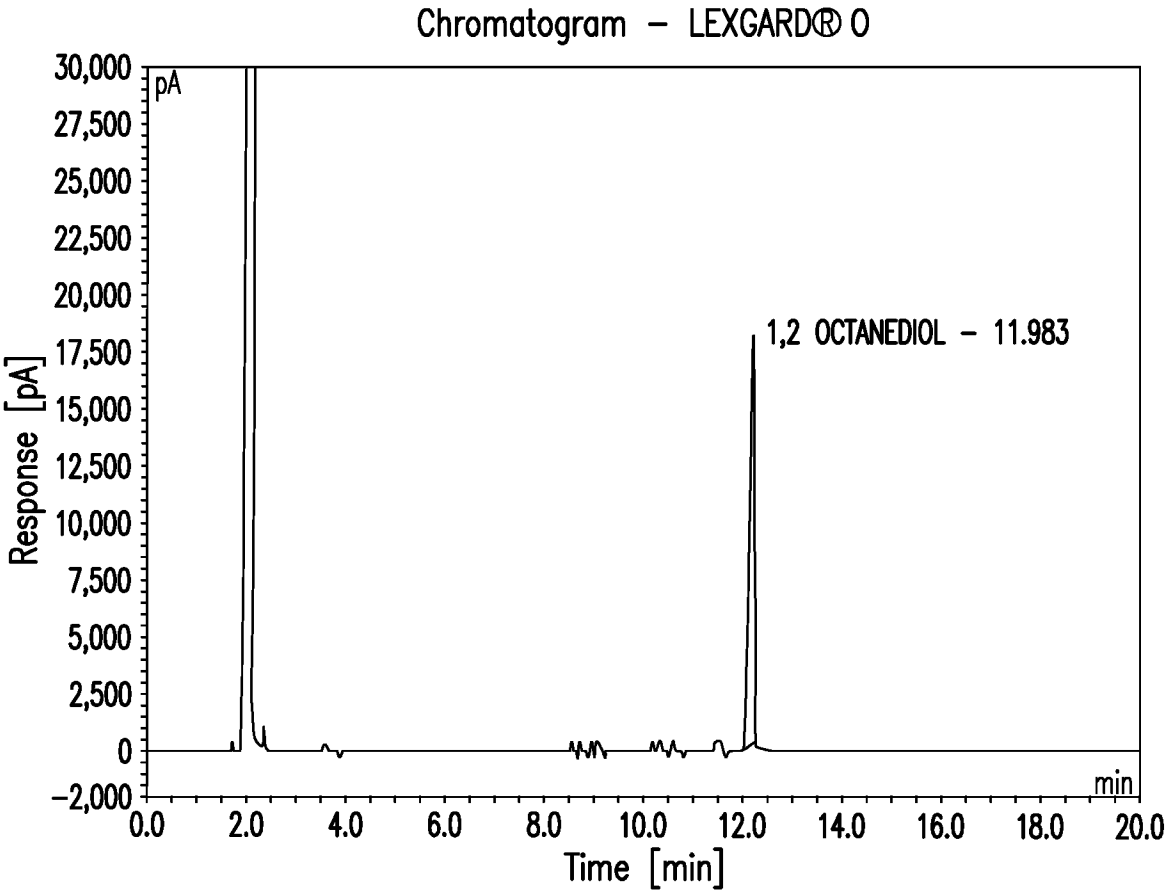

Chromatogram – LEXGARD® O
pA
1,2 OCTANEDIOL – 11.983
min
Response [pA]
Time [min]
30,000
27,500
25,000
22,500
20,000
17,500
15,000
12,500
10,000
7,500
5,000
2,500
0
−2,000
0.0
2.0
4.0
6.0
8.0
10.0
12.0
14.0
16.0
18.0
20.0

FIG.5

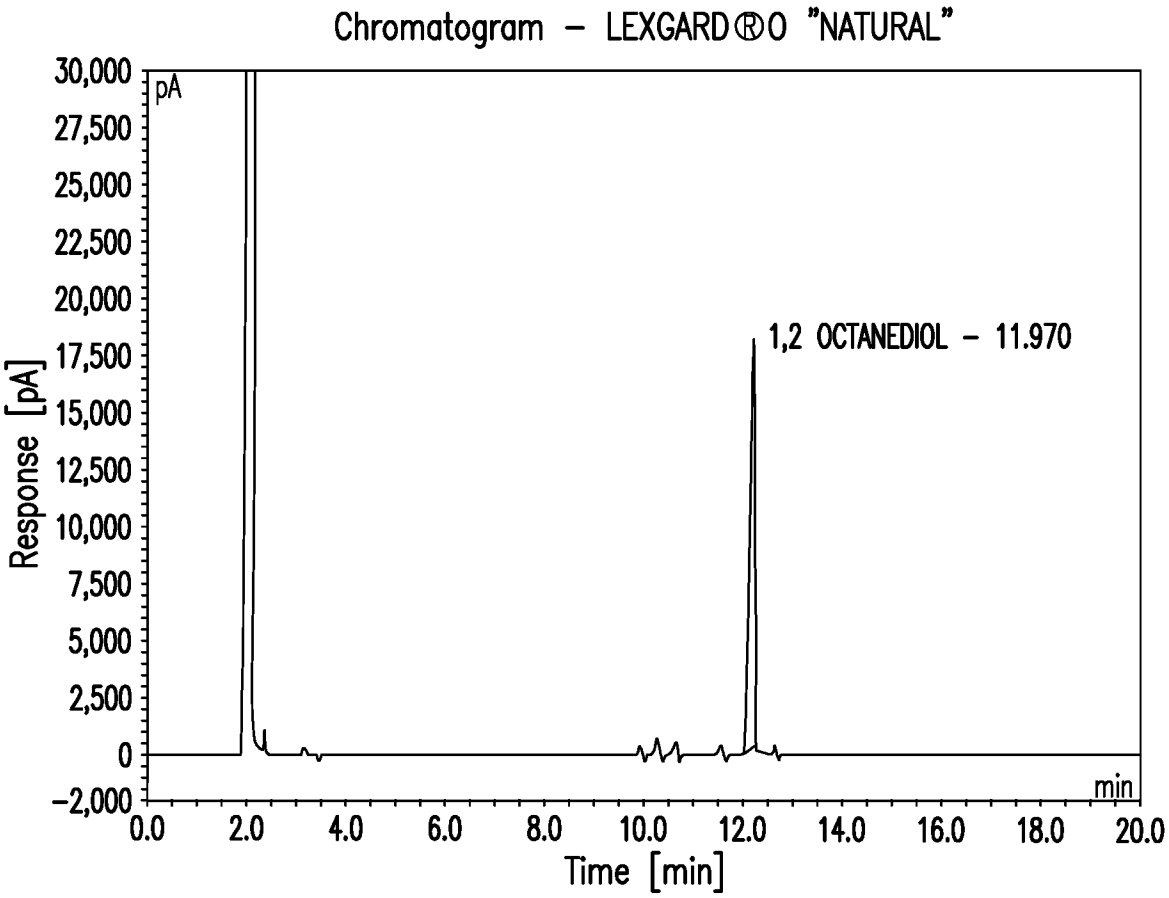

Chromatogram – LEXGARD®O "NATURAL"
pA
1,2 OCTANEDIOL – 11.970
min
Response [pA]
30,000
27,500
25,000
22,500
20,000
17,500
15,000
12,500
10,000
7,500
5,000
2,500
0
-2,000
0.0
2.0
4.0
6.0
8.0
10.0
12.0
14.0
16.0
18.0
20.0
Time [min]

FIG.6

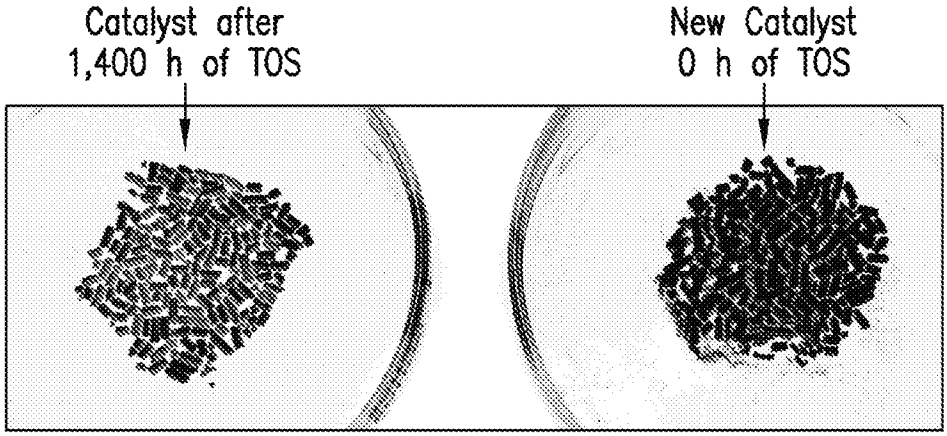


FIG.7

ELECTRONICALLY FILED

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Non-Provisional Patent Application of: Inolex Investment Corporation			
Conf. No.:	1028	Group Art Unit:	Not Yet Assigned
Appln. No.:	16/262,902	Examiner:	
Filing Date:	January 30, 2019	Attorney Docket No.:	I0113-198U2
Title:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making The Same.		

SECOND PRELIMINARY AMENDMENT IN U.S. NON-PROVISIONAL APPLICATION UNDER 37 C.F.R. 1.53(b)

Preliminary to the calculation of fees and examination of the above application being transmitted herewith, please amend the above-identified application as amended on the filing date in the first preliminary amendment, without prejudice, as follows:

Amendments to and Listing of the Claims:

Please cancel claims 56-71 and 81-87 without prejudice to the filing of one or more continuation or divisional applications so that the claims read as follows, with the changes as shown:

1.-41 (canceled)

42. (original) A composition, comprising at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

43. (original) The composition according to claim 42, wherein the composition is a personal care composition.

44. (original) The composition according to claim 43, wherein the personal care composition is a hair care composition, an oral care composition, a skin care composition or a cosmetic composition.

45. (original) The composition according to claim 42, wherein the composition is a composition for a household product.

46. (original) The composition according to claim 45, wherein the household product is a fabric care product or a cleaning product.

47. (original) The composition according to claim 42, wherein the composition is an industrial composition.

48. (original) The composition according to claim 42, wherein the composition is a pharmaceutical composition, a vitamin composition, or a health care composition.

49. (original) The composition according to claim 42, comprising a second bio-1,2-alkanediol, different than the first bio-1,2-alkanediol, the second bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a second bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

50. (original) The composition according to claim 49, wherein the composition further comprises at least one other bio-compound different from the first and the second bio-1,2-alkanediols.

51. (original) The composition according to claim 50, wherein the composition comprises an antimicrobial compound different from the first and the second bio-1,2-alkanediol and different from the other bio-compound.

52. (original) The composition according to claim 42, wherein the composition further comprises at least one other bio-compound different from the at least one bio-1,2-alkanediol.

53. (original) The composition according to claim 52, wherein the composition comprises at least one antimicrobial compound different from the at least one bio-1,2-alkanediol and different from the other bio-compound.

54. (original) The composition according to claim 42, wherein the first bio-1-alkene has a regioselectivity of about 95% to about 99%.

55. (original) The composition according to claim 42, wherein the first bio-alkene is bio-octene and the at least one first bio-1,2-alkane diol is bio-1,2-octanediol.

56. -71. (canceled)

72. (original) An antimicrobial product, comprising at least one first bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

73. (original) The antimicrobial product according to claim 72, further comprising a second bio-1,2-alkanediol, different than the at least one first bio-1,2-alkanediol, the second bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms and formed by conversion of a second bio-alkene, different than the first bio-alkene, having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

74. (original) The antimicrobial product according to claim 73, wherein the product further comprises at least one other bio-compound different from the at least one first and the second bio-1,2-alkanediols.

75. (original) The antimicrobial product according to claim 74, further comprising a second antimicrobial and/or a preservative different from the antimicrobial product.

76. (original) The antimicrobial product according to claim 72, wherein the product further comprises at least one other bio-compound different from the at least one first bio-1,2-alkanediol.

77. (original) The antimicrobial product according to claim 76, further comprising a second antimicrobial and/or a preservative different from the antimicrobial product.

78. (original) The antimicrobial product according to claim 72, wherein the first bio-1-alkene has a regioselectivity of about 95% to about 99%.

79. (original) The antimicrobial product according to claim 72, wherein the antimicrobial product demonstrates antimicrobial efficacy.

80. (original) The antimicrobial product according to claim 72, wherein the first bio-alkene is bio-octene and the at least one first 1,2-alkane diol is bio-1,2-octanediol.

81. -87 (canceled)

REMARKS

Claims 42-55 and 72-80 are presently pending in this application.

Claims 1-41 were canceled at the time of filing without prejudice to the filing of one or more divisional or continuation applications.

Claims 56-71 and 81-87 are canceled herein also without prejudice to the filing of one or more divisional or continuation applications.

No new matter has been added by these amendments, and entry thereof and an early notice of allowance are respectfully requested.

Respectfully submitted,

INOLEX INVESTMENT CORPORATION

6/19/19 By: _____
(Date)

LYNDA L. CALDERONE
Registration No. 35,837
CALDERONE BULLOCK LLC
Two Greentree Centre
9000 Lincoln Drive East, Suite 301
Marlton, NJ 08053
Telephone: 856-446-6047
Direct Dial: 845-446-6045
Facsimile: 856-267-5370
E-Mail: lcalderone@calderonebullock.com

LLC:llc

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NUMBER	FILING or 371(c) DATE	GRP ART UNIT	FIL FEE REC'D	ATTY.DOCKET.NO	TOT CLAIMS	IND CLAIMS
16/262,902	01/30/2019		2180	I0113-198U2	23	2

CONFIRMATION NO. 1028

UPDATED FILING RECEIPT

159443
Calderone Bullock LLC
Two Greentree Centre, Suite 301
9000 Lincoln Drive East
Marlton, NJ 08053

OC000000109082378

Date Mailed: 06/21/2019

Receipt is acknowledged of this non-provisional utility patent application. The application will be taken up for examination in due course. Applicant will be notified as to the results of the examination. Any correspondence concerning the application must include the following identification information: the U.S. APPLICATION NUMBER, FILING DATE, NAME OF FIRST INVENTOR, and TITLE OF INVENTION. Fees transmitted by check or draft are subject to collection.

Please verify the accuracy of the data presented on this receipt. If an error is noted on this Filing Receipt, please submit a written request for a corrected Filing Receipt, including a properly marked-up ADS showing the changes with strike-through for deletions and underlining for additions. If you received a "Notice to File Missing Parts" or other Notice requiring a response for this application, please submit any request for correction to this Filing Receipt with your reply to the Notice. When the USPTO processes the reply to the Notice, the USPTO will generate another Filing Receipt incorporating the requested corrections provided that the request is grantable.

Inventor(s)

Rocco V. Burgo, Mullica Hill, NJ;
Michael E. Wright, Hurricane, UT;
Gary B. Mosser, Tabernacle, NJ;
Michael J. Fevola, Belle Mead, NJ;

Applicant(s)

Inolex Investment Corporation, Wilmington, DE

Assignment For Published Patent Application

Inolex Investment Corporation

Power of Attorney: None

Domestic Priority data as claimed by applicant

This appln claims benefit of 62/623,985 01/30/2018

Foreign Applications for which priority is claimed (You may be eligible to benefit from the **Patent Prosecution Highway** program at the USPTO. Please see http://www.uspto.gov for more information.) - None.
Foreign application information must be provided in an Application Data Sheet in order to constitute a claim to foreign priority. See 37 CFR 1.55 and 1.76.

Permission to Access Application via Priority Document Exchange: Yes

Permission to Access Search Results: Yes

Applicant may provide or rescind an authorization for access using Form PTO/SB/39 or Form PTO/SB/69 as appropriate.

If Required, Foreign Filing License Granted: 02/15/2019

The country code and number of your priority application, to be used for filing abroad under the Paris Convention, is **US 16/262,902**

Projected Publication Date: 09/26/2019

Non-Publication Request: No

Early Publication Request: No

Title

Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same

Preliminary Class

Statement under 37 CFR 1.55 or 1.78 for AIA (First Inventor to File) Transition Applications: No

PROTECTING YOUR INVENTION OUTSIDE THE UNITED STATES

Since the rights granted by a U.S. patent extend only throughout the territory of the United States and have no effect in a foreign country, an inventor who wishes patent protection in another country must apply for a patent in a specific country or in regional patent offices. Applicants may wish to consider the filing of an international application under the Patent Cooperation Treaty (PCT). An international (PCT) application generally has the same effect as a regular national patent application in each PCT-member country. The PCT process **simplifies** the filing of patent applications on the same invention in member countries, but **does not result** in a grant of "an international patent" and does not eliminate the need of applicants to file additional documents and fees in countries where patent protection is desired.

Almost every country has its own patent law, and a person desiring a patent in a particular country must make an application for patent in that country in accordance with its particular laws. Since the laws of many countries differ in various respects from the patent law of the United States, applicants are advised to seek guidance from specific foreign countries to ensure that patent rights are not lost prematurely.

Applicants also are advised that in the case of inventions made in the United States, the Director of the USPTO must issue a license before applicants can apply for a patent in a foreign country. The filing of a U.S. patent application serves as a request for a foreign filing license. The application's filing receipt contains further information and guidance as to the status of applicant's license for foreign filing.

Applicants may wish to consult the USPTO booklet, "General Information Concerning Patents" (specifically, the section entitled "Treaties and Foreign Patents") for more information on timeframes and deadlines for filing foreign patent applications. The guide is available either by contacting the USPTO Contact Center at 800-786-9199, or it can be viewed on the USPTO website at http://www.uspto.gov/web/offices/pac/doc/general/index.html.

For information on preventing theft of your intellectual property (patents, trademarks and copyrights), you may wish to consult the U.S. Government website, http://www.stopfakes.gov. Part of a Department of Commerce initiative, this website includes self-help "toolkits" giving innovators guidance on how to protect intellectual property in specific

countries such as China, Korea and Mexico. For questions regarding patent enforcement issues, applicants may call the U.S. Government hotline at 1-866-999-HALT (1-866-999-4258).

LICENSE FOR FOREIGN FILING UNDER

Title 35, United States Code, Section 184

Title 37, Code of Federal Regulations, 5.11 & 5.15

GRANTED

The applicant has been granted a license under 35 U.S.C. 184, if the phrase "IF REQUIRED, FOREIGN FILING LICENSE GRANTED" followed by a date appears on this form. Such licenses are issued in all applications where the conditions for issuance of a license have been met, regardless of whether or not a license may be required as set forth in 37 CFR 5.15. The scope and limitations of this license are set forth in 37 CFR 5.15(a) unless an earlier license has been issued under 37 CFR 5.15(b). The license is subject to revocation upon written notification. The date indicated is the effective date of the license, unless an earlier license of similar scope has been granted under 37 CFR 5.13 or 5.14.

This license is to be retained by the licensee and may be used at any time on or after the effective date thereof unless it is revoked. This license is automatically transferred to any related applications(s) filed under 37 CFR 1.53(d). This license is not retroactive.

The grant of a license does not in any way lessen the responsibility of a licensee for the security of the subject matter as imposed by any Government contract or the provisions of existing laws relating to espionage and the national security or the export of technical data. Licensees should apprise themselves of current regulations especially with respect to certain countries, of other agencies, particularly the Office of Defense Trade Controls, Department of State (with respect to Arms, Munitions and Implements of War (22 CFR 121-128)); the Bureau of Industry and Security, Department of Commerce (15 CFR parts 730-774); the Office of Foreign AssetsControl, Department of Treasury (31 CFR Parts 500+) and the Department of Energy.

NOT GRANTED

No license under 35 U.S.C. 184 has been granted at this time, if the phrase "IF REQUIRED, FOREIGN FILING LICENSE GRANTED" DOES NOT appear on this form. Applicant may still petition for a license under 37 CFR 5.12, if a license is desired before the expiration of 6 months from the filing date of the application. If 6 months has lapsed from the filing date of this application and the licensee has not received any indication of a secrecy order under 35 U.S.C. 181, the licensee may foreign file the application pursuant to 37 CFR 5.15(b).

SelectUSA

The United States represents the largest, most dynamic marketplace in the world and is an unparalleled location for business investment, innovation, and commercialization of new technologies. The U.S. offers tremendous resources and advantages for those who invest and manufacture goods here. Through SelectUSA, our nation works to promote and facilitate business investment. SelectUSA provides information assistance to the international investor community; serves as an ombudsman for existing and potential investors; advocates on behalf of U.S. cities, states, and regions competing for global investment; and counsels U.S. economic development organizations on investment attraction best practices. To learn more about why the United States is the best country in the world to develop

technology, manufacture products, deliver services, and grow your business, visit http://www.SelectUSA.gov or call +1-202-482-6800.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NUMBER	FILING OR 371(C) DATE	FIRST NAMED APPLICANT	ATTY. DOCKET NO./TITLE
16/262,902	01/30/2019	Rocco V. Burgo	I0113-198U2

CONFIRMATION NO. 1028

159443
Calderone Bullock LLC
Two Greentree Centre, Suite 301
9000 Lincoln Drive East
Marlton, NJ 08053

INFORMAL NOTICE

OC00000109082379

Date Mailed: 06/21/2019

INFORMATIONAL NOTICE TO APPLICANT

Applicant is notified that the above-identified application contains the deficiencies noted below. No period for reply is set forth in this notice for correction of these deficiencies. However, if a deficiency relates to the inventor's oath or declaration, the applicant must file an oath or declaration in compliance with 37 CFR 1.63, or a substitute statement in compliance with 37 CFR 1.64, executed by or with respect to each actual inventor no later than the expiration of the time period set in the "Notice of Allowability" to avoid abandonment. See 37 CFR 1.53(f).

The item(s) indicated below are also required and should be submitted with any reply to this notice to avoid further processing delays.

- A properly executed inventor's oath or declaration has not been received for the following inventor(s):
  Rocco V. Burgo
  Michael E. Wright
  Gary B. Mosser
  Michael J. Fevola

Questions about the contents of this notice and the requirements it sets forth should be directed to the Office of Data Management, Application Assistance Unit, at **(571) 272-4000** or **(571) 272-4200** or **1-888-786-0101**.

/thaile/

PATENT APPLICATION FEE DETERMINATION RECORD

Substitute for Form PTO-875

Application or Docket Number: 16/262,902

APPLICATION AS FILED - PART I

FOR	(Column 1) NUMBER FILED	(Column 2) NUMBER EXTRA	SMALL ENTITY RATE($)	SMALL ENTITY FEE($)	OR	OTHER THAN SMALL ENTITY RATE($)	OTHER THAN SMALL ENTITY FEE($)
BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A	N/A			N/A	300
SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A	N/A			N/A	660
EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A	N/A			N/A	760
TOTAL CLAIMS (37 CFR 1.16(i))	23 minus 20 =	* 3			OR	x 100 =	300
INDEPENDENT CLAIMS (37 CFR 1.16(h))	2 minus 3 =	*				x 460 =	0.00
APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $310 ($155 for small entity) for each additional 50 sheets or fraction thereof. See 35 U.S.C. 41(a)(1)(G) and 37 CFR 1.16(s).						0.00
MULTIPLE DEPENDENT CLAIM PRESENT (37 CFR 1.16(j))							0.00
* If the difference in column 1 is less than zero, enter "0" in column 2.			TOTAL			TOTAL	2020

APPLICATION AS AMENDED - PART II

		(Column 1) CLAIMS REMAINING AFTER AMENDMENT		(Column 2) HIGHEST NUMBER PREVIOUSLY PAID FOR	(Column 3) PRESENT EXTRA	SMALL ENTITY RATE($)	SMALL ENTITY ADDITIONAL FEE($)	OR	OTHER THAN SMALL ENTITY RATE($)	OTHER THAN SMALL ENTITY ADDITIONAL FEE($)
AMENDMENT A	Total (37 CFR 1.16(i))	*	Minus	**	=	x =		OR	x =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=	x =		OR	x =	
	Application Size Fee (37 CFR 1.16(s))									
	FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							OR		
						TOTAL ADD'L FEE		OR	TOTAL ADD'L FEE	

		(Column 1) CLAIMS REMAINING AFTER AMENDMENT		(Column 2) HIGHEST NUMBER PREVIOUSLY PAID FOR	(Column 3) PRESENT EXTRA	SMALL ENTITY RATE($)	SMALL ENTITY ADDITIONAL FEE($)	OR	OTHER THAN SMALL ENTITY RATE($)	OTHER THAN SMALL ENTITY ADDITIONAL FEE($)
AMENDMENT B	Total (37 CFR 1.16(i))	*	Minus	**	=	x =		OR	x =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=	x =		OR	x =	
	Application Size Fee (37 CFR 1.16(s))									
	FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							OR		
						TOTAL ADD'L FEE		OR	TOTAL ADD'L FEE	

* If the entry in column 1 is less than the entry in column 2, write "0" in column 3.

** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 20, enter "20".

*** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 3, enter "3".

The "Highest Number Previously Paid For" (Total or Independent) is the highest found in the appropriate box in column 1.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NUMBER	FILING OR 371(C) DATE	FIRST NAMED APPLICANT	ATTY. DOCKET NO./TITLE
16/262,902	01/30/2019	Rocco V. Burgo	I0113-198U2

CONFIRMATION NO. 1028

159443
Calderone Bullock LLC
Two Greentree Centre, Suite 301
9000 Lincoln Drive East
Marlton, NJ 08053

PUBLICATION NOTICE

OC000000111457655

Title: Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same

Publication No.US-2019-0289848-A1
Publication Date:09/26/2019

NOTICE OF PUBLICATION OF APPLICATION

The above-identified application will be electronically published as a patent application publication pursuant to 37 CFR 1.211, et seq. The patent application publication number and publication date are set forth above.

The publication may be accessed through the USPTO's publically available Searchable Databases via the Internet at www.uspto.gov. The direct link to access the publication is currently http://www.uspto.gov/patft/.

The publication process established by the Office does not provide for mailing a copy of the publication to applicant. A copy of the publication may be obtained from the Office upon payment of the appropriate fee set forth in 37 CFR 1.19(a)(1). Orders for copies of patent application publications are handled by the USPTO's Public Records Division. The Public Records Division can be reached by telephone at (571) 272-3150 or (800) 972-6382, by facsimile at (571) 273-3250, by mail addressed to the United States Patent and Trademark Office, Public Records Division, Alexandria, VA 22313-1450 or via the Internet.

In addition, information on the status of the application, including the mailing date of Office actions and the dates of receipt of correspondence filed in the Office, may also be accessed via the Internet through the Patent Electronic Business Center at www.uspto.gov using the public side of the Patent Application Information and Retrieval (PAIR) system. The direct link to access this status information is currently https://portal.uspto.gov/pair/PublicPair. Prior to publication, such status information is confidential and may only be obtained by applicant using the private side of PAIR.

Further assistance in electronically accessing the publication, or about PAIR, is available by calling the Patent Electronic Business Center at 1-866-217-9197.

Office of Data Managment, Application Assistance Unit (571) 272-4000, or (571) 272-4200, or 1-888-786-0101

FILED ELECTRONICALLY

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Patent Application of: Inolex Investment Corporation			
Conf. No.:	1028	Group Art Unit:	1612
Appln. No.:	16/262,902	Examiner:	Nannette Holloman
Filing Date:	January 30, 2019	Attorney Docket No.:	I0113-198U2
Title:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same		

TRANSMITTAL OF DOCUMENTS

Enclosed are the following documents:

[X] [X] Inventor Declaration; [ ] Inventor(s) Declaration and Power of Attorney;] Inventor(s) Oath; [ ] Substitute Statement

[ ] Appointment of Attorney by Applicant/Assignee.

[ ] General Power of Attorney and 3.73(c) Statement

[ ] Revocation and Appointment of Attorney and Change of Correspondence Address.

[ ] Replacement Drawings.

[ ] Substitute Application Data Sheet.

[ ] Verified English translation of application as filed.

[ ] Priority document(s): Certified copy(ies) of __ patent application No(s). __, filed in __ on __ from which priority is claimed under 35 U.S.C. § 119.

[ ] Microfiche computer program (Appendix).

[ ] Nucleotide and/or Amino Acid Sequence Submission, including:

[ ] Computer readable copy; [ ] Paper Copy; [ ] Verified Statement.

[ ] [ ] Formal drawings: _____ sheets; [ ] Replacement drawings: ___ sheets

[ ] Information Disclosure Statement, PTO/SB/08A or B and cited references.

[ ] Request for Refund.

[ ] Request for Corrected Filing Receipt. Attached is a marked-up copy of the Filing Receipt showing corrections.

[ ] Applicant(s), by its/their undersigned attorney, claim(s) Small Entity Status under 37 C.F.R. §1.27 as [ ] an Independent Inventor, or [ ] a Small Business Concern, or [ ] a Non-Profit Organization.

[ ] Micro Entity Status [ ] is hereby claimed by Applicant(s) and Micro Entity Certification is enclosed; [ ] has previously been claimed/established.

[ ] Preliminary Amendment. [ ] The application filing fees were paid upon filing and additional claim fees resulting from the Preliminary Amendment are calculated as follows:

					MICRO ENTITY		SMALL ENTITY		LARGE ENTITY	
	NO. OF CLAIMS AFTER AMEND.		HIGHEST NO. PREVIOUSLY PAID FOR	EXTRA	RATE	ADDIT. FEE	RATE	ADDIT. FEE	RATE	ADDIT. FEE
TOTAL		-			X $25	$	X $50	$	X $100	
INDEP.		-			X $115	$	X$230	$	X $460	
[ ] 1st PRESENTATION OF MULTIPLE DEPENDENT CLAIMS					$205		$410		$820	
					TOTAL	$	TOTAL	$	TOTAL	

[ ] Utility Non-Provisional Patent Application Filing Fees. The application filing fees were not paid at filing and are being paid herewith and are calculated as follows:

				MICRO ENTITY			SMALL ENTITY			LARGE ENTITY	
CLAIMS	NO. FILED		NO. EXTRA	BASIC FILING, SEARCH, AND EXAMINATION FEES:	$430		BASIC FILING, SEARCH, AND EXAMINATION FEES:	$ 785		BASIC FILING, SEARCH, AND EXAMINATION FEES:	$1,720
Total	@% -20 =		@%	X $ 25	$	OR	X $ 50	$	OR	X $100	$
Independent	@% - 3=		@%	X $115	$	OR	X $230	$	OR	X $460	$
[@%] Multiple Dependent Claims				X $205	$	OR	X $410	$	OR	X $820	$
[ ] Application Size Fee (if specification and drawings exceed 100 sheets)											
Total sheets	-100	Add'l sheets =	# of sets of 50 (round up to whole number) /50 =	X $100 =	$	OR	X $200 =	$	OR	X $400	$
Late Fee For Filing Fees, Inventor Oath or Declaration				$ 40	$	OR	$ 80	$	OR	$160	$

Late Fee For Non-English Specification	$ 35	$	OR	$ 70	$	OR	$140	$
	TOTAL	$	OR	TOTAL	$	OR	TOTAL	$

[ ] Other:

[ ] Applicant(s) herewith charge the following fees in the total amount of $_____ as calculated above to the credit card account, Calderone Bullock _____(Billing No. @%.@%):

[ ] Additional Claim Fees After Preliminary Amendment $____.

[ ] Non-Provisional Utility Patent Application Filing Fees $_____.

[X] The Commissioner is hereby authorized to charge and/or credit Deposit Account No. 60-2689 (Billing No. I0113.0251) as noted below.

[ ] Any overpayments or deficiencies in the above-calculated fees.

[X] Any additional fees required under 37 C.F.R. § 1.16 and § 1.17.

[X] In the event that a Petition for Extension of Time is required during the prosecution of this application, but not submitted, please charge any extension fee under 37 C.F.R. § 1.136(a) to our Deposit Account noted above.

[X] Applicant believes there are no additional fees required with this submission. Should a fee be necessary, the Commissioner is hereby authorized to charge any fee or deficiency, or credit any overpayment to Deposit Account No. 60-2689 (Billing No. I0113.0251).

INOLEX INVESTMENT CORPORATION

2/24/20 By: _____
(Date)

LYNDA L. CALDERONE
Registration No. 35,837
CALDERONE BULLOCK LLC
Two Greentree Centre
9000 Lincoln Drive East, Suite 301
Marlton, NJ 08053
Telephone: 856-446-6047
Direct Dial: 856-446-6045
Facsimile: 856-267-5370
E-Mail: lcalderone@calderonebullock.com

[X] Customer Number or Bar Code Label: **000159443**

LLC:smc
Enclosures: Inventor's Declaration

DECLARATION
(Original application)

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am an original sole inventor (if only one name is listed below) or an original joint inventor (if plural names are listed below) of the subject matter that is claimed and for which a patent is sought on the invention entitled:

NATURAL 1,2-ALKANEDIOLS, COMPOSITIONS HAVING NATURAL 1,2-ALKANEDIOLS AND PROCESSES FOR MAKING THE SAME.

the specification of which is attached hereto and/or was filed on January 30, 2019 as Application No. 16/262,902.

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to herein.

The above-identified application was made or was authorized to be made by me.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d), of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

FOREIGN PRIORITY APPLICATION(S)

(Number)	(Country)	(Day/month/year filed)	Priority Claimed [ ] Yes [ ] No
(Number)	(Country)	(Day/month/year filed)	[ ]Yes [ ] No

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional patent application(s) listed below:

PROVISIONAL PRIORITY PATENT APPLICATION(S)

		Priority Claimed
62/623,985	January 30, 2018	[X] Yes [ ] No
(Application No.)	(Filing Date)	
		[ ] Yes [ ] No
(Application No.)	(Filing Date)	

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements made in this Declaration are punishable under 18 U.S.C. §1001 by fine or imprisonment of not more than five (5) years.

Full name of first or sole inventor: Rocco V. Burgo

Inventor's Signature:

Date: 2/5/20

Residence: Mullica Hill, New Jersey

Citizenship: United States

Post Office Address: 604 Widgeon Court, Mullica Hill, New Jersey 08062

Full name of second inventor: Michael E. Wright

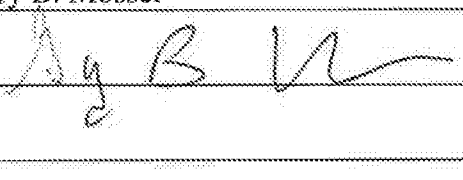
Inventor's Signature:

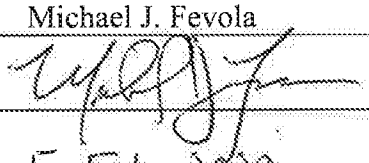
Date: 2/10/2020

Residence: Hurricane, Utah

Citizenship: United States

Post Office Address: 4175 S. 1200 W., Hurricane, Utah 84737

Full name of third inventor	Gary B. Mosser
Inventor's Signature	
Date	2/10/20
Residence	Tabernacle, New Jersey
Citizenship	United States
Post Office Address	5 Colonial Court, Tabernacle, New Jersey 08088

Full name of fourth inventor	Michael J. Fevola
Inventor's Signature	
Date	5 Feb 2020
Residence	Belle Mead, New Jersey
Citizenship	United States
Post Office Address	409 Township Line Road, Belle Mead, New Jersey 08502

Electronic Acknowledgement Receipt

EFS ID:	38699322
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	159443
Filer:	lynda Leigh Calderone
Filer Authorized By:	
Attorney Docket Number:	I0113-198U2
Receipt Date:	26-FEB-2020
Filing Date:	30-JAN-2019
Time Stamp:	20:25:36
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Transmittal Letter	transmittalofdec.pdf	166037 7d224b0ec0bcb79826b4ce5967ee54effaefbf05	no	3

Warnings:

Information:					
2	Oath or Declaration filed	declaration.pdf	126797 8bbe750c8b95ce22e3e3f5455c176e12a5e977a8	no	3
Warnings:					
Information:					
			Total Files Size (in bytes):	292834	

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

FILED ELECTRONICALLY

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Patent Applications of:
Inolex Investment Corporation
103 Springer Building
411 Silverside Road
Wilmington, DE 19810

POWER OF ATTORNEY TO PROSECUTE APPLICATIONS BEFORE THE U.S. PATENT AND TRADEMARK OFFICE

Inolex Investment Corporation, assignee of the above application, by its undersigned representative, hereby revokes all previous powers of attorney given in the application identified in the enclosed Statement Under 37 C.F.R. §3.73(c).

Inolex Investment Corporation, by its undersigned representative, hereby appoints the registered attorneys and agents associated with Customer No. 159443, Calderone Bullock LLC, as its attorneys or agents to represent the assignee before the United States Patent and Trademark Office (U.S. PTO) in connection with any and all patent applications assigned only to the undersigned according to the U.S. PTO assignment records or assignment documents attached to this form and/or to the enclosed Statement Under 37 C.F.R. §3.73(c) and in accordance therewith, with full power of substitution and revocation, to prosecute the application identified in the enclosed Statement Under 37 C.F.R. § 3.73(c) and to transact all business in the U.S. PTO connected therewith.

Please change the correspondence address for the application identified in the enclosed Statement Under 3.73(c) to the address associated with **Customer No. 159443, namely, CALDERONE BULLOCK LLC**, Two Greentree Centre, 9000 Lincoln Drive East, Marlton New Jersey 08053. Please direct all communications and telephone calls to Lynda Calderone at (856) 446-6045 (telephone) or (856) 267-5370 (facsimile).

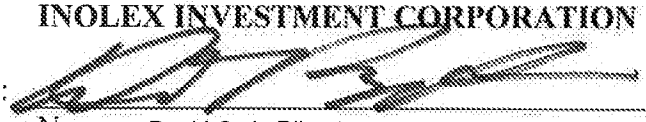
The undersigned states that he/she is authorized to sign the document on behalf of the assignee of the above application, and he/she certifies that, to the best of assignee's knowledge

and belief, title to the above application is in assignee by virtue of the documents provided and/or referenced in the enclosed Statement Under 37 C.F.R. §3.73(c).

Respectfully submitted,

INOLEX INVESTMENT CORPORATION

February 6, 2019
Date

By: 

Name: David C. A. Plimpton
Title: CEO
Telephone: 215-320-1507

Electronic Acknowledgement Receipt

EFS ID:	38864154
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	159443
Filer:	lynda Leigh Calderone
Filer Authorized By:	
Attorney Docket Number:	I0113-198U2
Receipt Date:	17-MAR-2020
Filing Date:	30-JAN-2019
Time Stamp:	18:13:13
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Assignee showing of ownership per 37 CFR 3.73	198U2373c.pdf	70393 bd8181d44dbbcc89cadcbaee16df9cecffbaa0a0	no	2

Warnings:

Information:					
2	Power of Attorney	POA.pdf	244247 183e9e56fe34e63561b7626aafae3f5370927726	no	2
Warnings:					
Information:					
			Total Files Size (in bytes):	314640	

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

ELECTRONICALLY FILED

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Patent Application of: Inolex Investment Corporation			
Conf. No.:	1028	Group Art Unit:	1612
Appln. No.:	16/262,902	Examiner:	Nannette Holloman
Filing Date:	January 30, 2019	Attorney Docket No.:	I0113-198U2

Title: Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same

STATEMENT UNDER 37 C.F.R. § 3.73(c)

3.73 (c) Statement:

Assignee, Inolex Investment Corporation, is owner of the entire right, title and interest in and to the above-referenced application and its related applications by virtue of: (a) the chain of title noted below wherein the documents are recorded as noted under 37 C.F.R. 3.73(c)(1)(ii) and in the Appointment of Attorney filed simultaneously herewith; or (b) the enclosed copies of the documentary evidence of the chain of title set forth below and in the Appointment of Attorney that, as required under 3.73(c)(1)(i), were submitted for recordation or are being concurrently submitted for recordation pursuant to 37 C.F.R. 11.

Inolex Investment Corporation is the owner by virtue of and as set forth in the Appointment of Attorney by Assignee filed simultaneously herewith:

An assignment from the inventors of the above-identified patent application dated February 17, 2020, February 20, 2020 and February 23, 2020. The assignment was recorded in the United States Patent and Trademark Office (PTO) on March 2, 2020 at Reel 051980, beginning at Frame 0482.

The undersigned (whose title appears below) is authorized to act on behalf of assignee in this application as set forth in the Power of Attorney by Assignee filed herewith.

Respectfully submitted,

INOLEX INVESTMENT CORPORATION

8/13/20 By: _____

Lynda L. Calderone, Managing Member
and Attorney for Assignee
Registration No. 35,837
Two Greentree Centre
9000 Lincoln Drive East, Suite 301
Marlton, NJ 08053
Telephone: 856-446-6047
Direct Dial: 856-446-6045
Facsimile: 856-267-5370
E-Mail: lcalderone@calderonebullock.com

LLC:smc
Enclosures
[X] Customer Number or Bar Code Label: **159443**

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NUMBER	FILING OR 371(C) DATE	FIRST NAMED APPLICANT	ATTY. DOCKET NO./TITLE
16/262,902	01/30/2019	Rocco V. Burgo	I0113-198U2

CONFIRMATION NO. 1028

POA ACCEPTANCE LETTER

159443
Calderone Bullock LLC
Two Greentree Centre, Suite 301
9000 Lincoln Drive East
Marlton, NJ 08053

OC000000115556673

Date Mailed: 03/20/2020

NOTICE OF ACCEPTANCE OF POWER OF ATTORNEY

This is in response to the Power of Attorney filed 03/17/2020.

The Power of Attorney in this application is accepted. Correspondence in this application will be mailed to the above address as provided by 37 CFR 1.33.

Questions about the contents of this notice and the requirements it sets forth should be directed to the Office of Data Management, Application Assistance Unit, at **(571) 272-4000** or **(571) 272-4200** or **1-888-786-0101**.

/nbekele/

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290
CONCISE DESCRIPTION OF RELEVANCE

Application Number	16262902

U.S. PATENTS

Cite No	Patent Number	Concise Description of Relevance

U.S. PATENT APPLICATION PUBLICATION

Cite No	Publication Number	Concise Description of Relevance
1	20140066666	See Attached

FOREIGN PATENT DOCUMENTS		
CiteNo	**Foreign Document Number**	**Concise Description of Relevance**

NON-PATENT PUBLICATIONS		
Cite No	**Reference**	**Concise Description of Relevance**
1	Jong & Birmingham, Mushrooms as a Source of Natural Flavor and AromaCcompounds", Chapter 37 (pp. 345-366) in MUSHROOM BIOLOGY AND MUSHROOM PRODUCTS (Chinese University Press 1993)	See attached.

2	Wang et al., Efficient conversion of microcrystalline cellulose to 1,2-alkanediols over supported Ni catalysts, GREEN CHEM., 14:758-765 (2012)	See attached.

PTO/SB/429(08-12)
Approved for use through 07/31/2015. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

U.S. PATENTS

Cite No	Patent Number	Kind Code[1]	Issue Date (YYYY-MM-DD)	First Named Inventor

U.S. PATENT APPLICATION PUBLICATIONS

Cite No	Publication Number	Kind Code[1]	Publication Date (YYYY-MM-DD)	First Named Inventor
1	20140066666	A1	2014-03-06	Oskar Koch

FOREIGN PATENTS AND PUBLISHED FOREIGN PATENT APPLICATIONS

Cite No	Foreign Document Number[3]	Country Code[2]	Kind Code[1]	Publication Date (YYYY-MM-DD)	Applicant, Patentee or First Named Inventor	T[5]
						☐

NON-PATENT PUBLICATIONS (e.g., journal article, Office action)

Cite No	Author (if any), title of the publication, page(s) being submitted, publication date, publisher (where available), place of publication (where available).	T[5]	E[6]

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

1	Wang et al., Efficient conversion of microcrystalline cellulose to 1,2-alkanediols over supported Ni catalysts, GREEN CHEM., 14:758-765 (2012)	☐	☐
2	Jong & Birmingham, Mushrooms as a Source of Natural Flavor and AromaCcompounds", Chapter 37 (pp. 345-366) in MUSHROOM BIOLOGY AND MUSHROOM PRODUCTS (Chinese University Press 1993)	☐	☐

STATEMENTS

The party making the submission is not an individual who has a duty to disclose information with respect to the above-identified application under 37 CFR 1.56.

This submission complies with the requirements of 35 U.S.C. 122(e) and 37 CFR 1.290.

☐ The fee set forth in 37 CFR 1.290(f) has been submitted herewith.

☒ The fee set forth in 37 CFR 1.290(f) is not required because this submission lists three or fewer total items and, to the knowledge of the person signing the statement after making reasonable inquiry, this submission is the first and the only submission under 35 U.S.C 122(e) filed in the above-identified application by the party making the submission or by a party in privity with the party.

☐ This resubmission is being made responsive to a notification of non-compliance issued for an earlier filed third-party submission. The corrections in this resubmission are limited to addressing the non-compliance. As such, the party making this resubmission: (1) requests that the Office apply the previously-paid fee set forth in 37 CFR 1.290(f), or (2) states that no fee is required to accompany this resubmission as the undersigned is again making the fee exemption statement set forth in 37 CFR 1.290(g).

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

Signature	/James Balls/		
Name/Print	James Balls	Registration Number (if applicable)	57703

Examiner Signature		Date Considered	

*EXAMINER: Signature indicates all documents listed above have been considered,except for citations through which a line is drawn. Draw line through citation if not considered. Include a copy of this form with next communication to applicant. 1. If known, enter kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16. See MPEP 901.04(a). 2. Enter the country or patent office that issued the document, by two-letter code under WIPO standard ST.3. See MPEP 1851. 3. For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. 4. If known, enter the kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 . See MPEP 901.04(a). 5. Check mark indicates translation attached. 6. Check mark indicates evidence of publication attached.

Third Party Submission

U.S. Serial No.16/262,902 (US2019/0289848)

Cite No.	Reference	Concise Description of Relevance
1	Wang *et al.*, *Efficient conversion of microcrystalline cellulose to 1,2-alkanediols over supported Ni catalysts*, GREEN CHEM., 14:758-765 (2012)	Wang describes 1,2-alkanediols according to claim 1 of US2019/0289848 (US 16/262,902). The 1,2-alkanediols of Wang are synthesized from crystalline cellulose as starting material. *See* abstract. Wang explains that cellulose is "the most abundant non-food biomass resource on earth, [and] presents a promising substitute for fossil fuel resources for the sustainable production of commodity chemicals and fuels which holds the possibility for carbon neutral technologies." *See* page 758, first col., second paragraph. Accordingly, the 1,2-alkanediols of Wang are non-petrochemically-derived, *i.e.*, they are "bio"-1,2-alkanediols according to claim 1 of US2019/0289848 (US 16/262,902). *See* paragraph [0012] of US2019/0289848.
2	Koch et al., US2014/0066666 (March 6, 2014)	Koch et al. describe a 1,2-alkanediol according to claim 1 of US2019/0289848 (US 16/262,902). Specifically, Koch et al. describes 1,2-pentandediol that is synthesized from furfuryl alcohol as starting material. *See* abstract. Koch et al. explain that furfuryl alcohol can be obtained in large amounts from renewable raw materials such as "sugar-containing cereal waste." *See* para. [0011]. An object of Koch et al. is to produce 1,2-pentanediol from environmentally friendly and/or protective of resources. *See* para. [0016]. Accordingly, the 1,2-pentandediol of Koch is non-petrochemically-derived, *i.e.*, it is "bio"-1,2-pentandediol according to claim 1 of US2019/0289848 (US 16/262,902). *See* paragraph [0012] of US2019/0289848.
3	Jong & Birmingham, *Mushrooms as a Source of Natural Flavor and AromaCcompounds*", Chapter 37 (pp. 345-366) in MUSHROOM BIOLOGY AND MUSHROOM PRODUCTS (Chinese University Press 1993)	Jong & Birmingham describe a 1,2-alkanediol according to claim 1 of US2019/0289848 (US 16/262,902). Jong & Birmingham report that 1,2-heptanediol is found in mushrooms. *See* p. 352 (Table 2) and p. 350 (explaining, "Table 2 lists aromatic compounds detected in mushrooms and their characteristic odors"). According to Jong & Birmingham, many fungi are capable of *de novo* synthesis of flavoring compounds (such as 1,2-heptanediol) when grown on standard culture media. *See* page 345, last two lines. Accordingly, the 1,2-heptanediol of Jong & Birmingham is non-petrochemically-derived, *i.e.*, it is "bio"-1,2-heptanediol according to claim 1 of US2019/0289848 (US 16/262,902). *See* paragraph [0012] of US2019/0289848.

Electronic Acknowledgement Receipt

EFS ID:	38975209
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	
First Named Inventor/Applicant Name:	
Correspondence Address:	- - - - - - - - - -
Filer:	James Balls
Filer Authorized By:	
Attorney Docket Number:	
Receipt Date:	26-MAR-2020
Filing Date:	
Time Stamp:	12:26:39
Application Type:	

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Concise Description of Relevance	Concise-description-generated.pdf	31502 0ac4a3d5de12551ab8fa83445a931a12687f830f	no	3
Warnings:					
Information:					
2	Third-Party Submission Under 37 CFR 1.290	Third-party-preissuance-submission.pdf	44018 80b7c34c27a9b2dded14ea6d9f05930abba7c41f	no	3
Warnings:					
Information:					
3	Request for Notification of Non-compliant Third-Party Submission	Third-party-notification-request.pdf	19656 e5ec1674c25f040e25f3a27115d722b9e4859c91	no	1
Warnings:					
Information:					
4	Concise Description of Relevance	Concise_Description.pdf	6682 05f47d3892c5997fd78be24a7faeb53963bbc396	no	1
Warnings:					
Information:					
5	Non Patent Literature	Wang.pdf	414639 d585b1fe3557e7187f7e93c4ab267f5850d5cf5b	no	8
Warnings:					
Information:					
6	Non Patent Literature	Jong.pdf	1263870 a8c80364c6250cad3cb30f6b24714736fe22ca54	no	13
Warnings:					
Information:					
		Total Files Size (in bytes):	1780367		

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	I0113-198U2	1028

159443 7590 04/02/2020

Calderone Bullock LLC
Two Greentree Centre, Suite 301
9000 Lincoln Drive East
Marlton, NJ 08053

EXAMINER	
HOLLOMAN, NANNETTE	

ART UNIT	PAPER NUMBER
1612	

NOTIFICATION DATE	DELIVERY MODE
04/02/2020	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

USP@calderonebullock.com
docketing@calderonebullock.com
kbullock@calderonebullock.com

UNITED STATES DEPARTMENT OF COMMERCE
U.S. Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450

APPLICATION NO./ CONTROL NO.	FILING DATE	FIRST NAMED INVENTOR/ PATENT IN REEXAMINATION	ATTORNEY DOCKET NO.
16/262,902	01/30/2019	Burgo et al.	I0113-198U2

		EXAMINER	
Calderone Bullock LLC Two Greentree Centre, Suite 301 9000 Lincoln Drive East Marlton, NJ 08053		MARIANNE C SEIDEL	
		ART UNIT	PAPER
		1600	20200331

DATE MAILED: _____

Please find below and/or attached an Office communication concerning this application or proceeding.

Commissioner for Patents

The third-party submission under 37 CFR 1.290 filed on 3/26/2020 for the instant application has been determined to be compliant with 35 U.S.C. 122(e) and 37 CFR 1.290 and is being entered in the application. Please allow a few days for the submission to be visible in the Patent Application Information Retrieval (PAIR) system.

/MARIANNE C SEIDEL/
Quality Assurance Specialist, Art Unit 1600

PTO-90C (Rev.04-03)

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290
CONCISE DESCRIPTION OF RELEVANCE

Application Number	16262902

U.S. PATENTS

Cite No	Patent Number	Concise Description of Relevance
1	4234752	See Attached

U.S. PATENT APPLICATION PUBLICATION

Cite No	Publication Number	Concise Description of Relevance
1	20060161025	See Attached

2	20030065233	See Attached
3	20050065389	See Attached

FOREIGN PATENT DOCUMENTS		
CiteNo	**Foreign Document Number**	**Concise Description of Relevance**
1	1876162	See Attached

NON-PATENT PUBLICATIONS		
Cite No	**Reference**	**Concise Description of Relevance**
1	Swern et al., Hydroxylation and Epoxidation of Some 1-Olefins with Per-acids, JACS, 68:1504-1507 (1946)	See Attached

PTO/SB/429(08-12)
Approved for use through 07/31/2015. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

U.S. PATENTS				
Cite No	Patent Number	Kind Code[1]	Issue Date (YYYY-MM-DD)	First Named Inventor
1	4234752		1980-11-18	YULIN WU

U.S. PATENT APPLICATION PUBLICATIONS				
Cite No	Publication Number	Kind Code[1]	Publication Date (YYYY-MM-DD)	First Named Inventor
1	20050065389	A1	2005-03-24	De Bruyn
2	20030065233	A1	2003-04-03	Fuji
3	20060161025	A1	2008-06-10	Swern

FOREIGN PATENTS AND PUBLISHED FOREIGN PATENT APPLICATIONS						
Cite No	Foreign Document Number[3]	Country Code[2]	Kind Code[1]	Publication Date (YYYY-MM-DD)	Applicant, Patentee or First Named Inventor	T[5]
1	1876162	EP	A1	2008-01-09	Furuya	☐

NON-PATENT PUBLICATIONS (e.g., journal article, Office action)

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

Cite No	Author (if any), title of the publication, page(s) being submitted, publication date, publisher (where available), place of publication (where available).	T[5]	E[6]
1	Swern et al., Hydroxylation and Epoxidation of Some 1-Olefins with Per-acids, JACS, 68:1504-1507 (1946)	☐	☐

STATEMENTS

The party making the submission is not an individual who has a duty to disclose information with respect to the above-identified application under 37 CFR 1.56.

This submission complies with the requirements of 35 U.S.C. 122(e) and 37 CFR 1.290.

☒ The fee set forth in 37 CFR 1.290(f) has been submitted herewith.

☐ The fee set forth in 37 CFR 1.290(f) is not required because this submission lists three or fewer total items and, to the knowledge of the person signing the statement after making reasonable inquiry, this submission is the first and the only submission under 35 U.S.C 122(e) filed in the above-identified application by the party making the submission or by a party in privity with the party.

☐ This resubmission is being made responsive to a notification of non-compliance issued for an earlier filed third-party submission. The corrections in this resubmission are limited to addressing the non-compliance. As such, the party making this resubmission: (1) requests that the Office apply the previously-paid fee set forth in 37 CFR 1.290(f), or (2) states that no fee is required to accompany this resubmission as the undersigned is again making the fee exemption statement set forth in 37 CFR 1.290(g).

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

Signature	/daniel altman/		
Name/Print	Daniel Altman	Registration Number (if applicable)	34115

Examiner Signature		Date Considered	

*EXAMINER: Signature indicates all documents listed above have been considered,except for citations through which a line is drawn. Draw line through citation if not considered. Include a copy of this form with next communication to applicant. 1. If known, enter kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16. See MPEP 901.04(a). 2. Enter the country or patent office that issued the document, by two-letter code under WIPO standard ST.3. See MPEP 1851. 3. For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. 4. If known, enter the kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 . See MPEP 901.04(a). 5. Check mark indicates translation attached. 6. Check mark indicates evidence of publication attached.

(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) EP 1 876 162 A1

(12) **EUROPEAN PATENT APPLICATION**
published in accordance with Art. 153(4) EPC

(43) Date of publication:
09.01.2008 Bulletin 2008/02

(21) Application number: **06732312.1**

(22) Date of filing: **25.04.2006**

(51) Int Cl.:
C07C 31/20 *(2006.01)*

(86) International application number:
PCT/JP2006/308652

(87) International publication number:
WO 2006/118111 (09.11.2006 Gazette 2006/45)

(84) Designated Contracting States:
DE FR GB

(30) Priority: **28.04.2005 JP 2005133200**

(71) Applicant: **Adeka Corporation**
Arakawa-ku
Tokyo 116-0012 (JP)

(72) Inventors:
- **FURUYA, Masaki, c/o ASAHI DENKA Co., Ltd.**
Tokyo 116-0012 (JP)
- **TSUSHIMA, Yasuhiro, c/o ASAHI DENKA Co., Ltd.**
Tokyo 116-0012 (JP)
- **NAMIWA, Kimiyoshi, c/o ASAHI DENKA Co., Ltd.**
Tokyo 116-0012 (JP)

(74) Representative: **Jones, Helen M.M. et al**
Gill Jennings & Every LLP
Broadgate House
7 Eldon Street
London EC2M 7LH (GB)

(54) ALKANEDIOL COMPOSITION, PROCESS FOR PRODUCING THE SAME AND COSMETIC

(57) A less malodorous alkanediol composition, a process for producing the alkanediol composition efficiently, and a cosmetic containing the alkanediol composition are provided.

An alkanediol composition contains 0.005 parts by mass or less of ester compound per 100 parts by mass of alkanediol compound having four or more carbon atoms. An alkanediol composition contains 0.2 parts by mass or less of dioxane compound per 100 parts by mass of alkanediol compound having four or more carbon atoms. Furthermore, an ether-containing dihydric alcohol is preferably 0.3 parts by mass or less per 100 parts by mass of alkanediol compound.

Printed by Jouve, 75001 PARIS (FR)

Description

Technical Field

[0001] The present invention relates to an alkanediol composition, a process for producing the alkanediol composition, and a cosmetic, and more particularly to an alkanediol composition that contains little specific by-products and that generates no unpleasant smell, a process for producing the alkanediol composition, and a cosmetic.

Background Art

[0002] It is unavoidable for manufacturers of organic compounds that organic chemical reactions are associated with the formation of by-products. The amount of by-product is often much smaller than that of end product. However, even a small amount of by-product sometimes chemically affects an end product, for example, by worsening the color or smell of the end product. For manufacturers of organic compounds, preventing the formation of disadvantageous by-products or finally removing disadvantageous by-products to prevent these adverse effects is part of manufacturing processes.

[0003] In such a situation, alkanediol compounds have been used in various applications, such as cosmetic preservatives and humectants, pearling agents, raw materials for synthetic fiber, raw materials for urethane compounds, and water-based inks. However, particularly in cosmetic applications, the smell of alkanediol compounds formulated in products causes a big problem. For example, the addition of an alkanediol compound to a product free from a perfume impairs the unscented product. Even in a product containing a perfume, the addition of a malodorous alkanediol compound changes the smell of the product. Hence, there has been a great demand for an odorless or less malodorous alkanediol composition in cosmetic applications.

[0004] A currently common process for producing an alkanediol composition includes oxidizing an olefin with an oxidizing agent such as hydrogen peroxide to produce an epoxy compound and hydrolyzing the epoxy compound. Because an alkanediol composition thus produced has a smell, the alkanediol composition is usually purified by distillation, which is a common purification method (see Patent Documents 1 and 2, for example).

[0005] Patent Document 1: Japanese Unexamined Patent Application Publication No. 57-62234

[0006] Patent Document 2: Japanese Unexamined Patent Application Publication No. 60-78928

Disclosure of Invention

Problems to be Solved by the Invention

[0007] However, as described in Patent Document 1 or 2, alkanediol compositions have a smell even after distillation. This is because a component causing a smell is not identified, and the distillation is performed without knowing the component to be removed. Accordingly, it is an object of the present invention to provide a less malodorous alkanediol composition, a process for producing the alkanediol composition efficiently, and a cosmetic containing the alkanediol composition.

Means for Solving the Problems

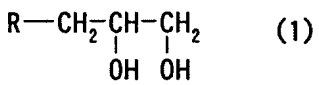
[0008] As a result of diligent investigations on the above-mentioned problems, the present inventors identified a specific component causing a smell in an alkanediol composition and perfected the present invention by discovering that a less malodorous alkanediol composition can be produced by reducing the specific compound to a certain level.

[0009] Thus, an alkanediol composition according to the present invention contains 0.005 parts by mass or less of ester compound (A) per 100 parts by mass of alkanediol compound having four or more carbon atoms.

[0010] Furthermore, another alkanediol composition according to the present invention contains 0.2 parts by mass or less of dioxane compound (C) per 100 parts by mass of alkanediol compound having four or more carbon atoms.

[0011] Furthermore, in the present invention, an ether-containing dihydric alcohol (B) is preferably 0.3 parts by mass or less per 100 parts by mass of alkanediol compound.

[0012] Furthermore, an alkanediol compound having a general formula (1) can suitably be used in the present invention:



(1)

wherein R denotes an alkyl group having 4-15 carbon atoms. Furthermore, the alkanediol compound is preferably a hydrolysate of an epoxy compound produced by an oxidation reaction of an olefin.

[0013] Furthermore, a process for producing an alkanediol composition according to the present invention includes the steps of (a) oxidizing an olefin to produce an epoxy compound, (b) hydrolyzing the epoxy compound to produce the alkanediol composition, and (c) adding water and/or an organic solvent to the alkanediol composition and removing water and/or the organic solvent under reduced pressure.

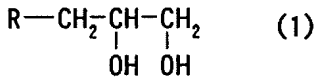
[0014] A cosmetic according to the present invention contains an alkanediol composition according to the present invention. Advantages of the Invention

[0015] An alkanediol composition according to the present invention has little smell. Furthermore, cosmetics containing an alkanediol composition according to the present invention do not have any problem caused by the smell of the alkanediol composition. An alkanediol composition containing a low concentration of component causing a smell can efficiently be produced by a process for producing an alkanediol composition according to the present invention.

Best Mode for Carrying Out the Invention

[0016] An alkanediol compound that can be used in the present invention has at least four carbon atoms. In addition, any two hydrogen atoms of a saturated aliphatic hydrocarbon of the alkanediol compound are replaced by hydroxyl groups. Examples of the alkanediol compound include 1,2-butanediol, 1,3-butanediol, 1,4-butanediol, 1,5-pentanediol, neopentyl glycol, isoprene glycol (3-methyl-1,3-butanediol), 1,2-hexanediol, 1,6-hexanediol, 3-methyl-1,5-pentanediol, 1,2-octanediol, 2,3-octanediol, 2-ethyl-1,3-hexanediol, 2-butyl-2-ethyl-1,3-propanediol, 2,5-dimethyl-2,5-hexanediol, 1,2-decanediol, 1,2-dodecanediol, 1,2-tetradecanediol, 1,2-hexadecanediol, 1,2-octadecanediol, 1,12-octadecanediol, 1,2-cyclohexanediol, 1,4-cyclohexanediol, and 1,4-cyclohexanedimethanol.

[0017] Among others, vicinal compounds that have two hydroxyl groups on adjacent carbon atoms are preferred. Compounds having a general formula (1) are more preferred:



(1)

wherein R denotes an alkyl group having 4-15 carbon atoms. Examples of the alkyl group include a butyl group, an isobutyl group, a secondary butyl group, a tertiary butyl group, a pentyl group, an isopentyl group, a secondary pentyl group, a neopentyl group, a tertiary pentyl group, a hexyl group, a secondary hexyl group, a heptyl group, a secondary heptyl group, an octyl group, a secondary octyl group, a nonyl group, a secondary nonyl group, a decyl group, a secondary decyl group, an undecyl group, a secondary undecyl group, a dodecyl group, a secondary dodecyl group, a tridecyl group, an isotridecyl group, a secondary tridecyl group, a tetradecyl group, a secondary tetradecyl group, a hexadecyl group, a secondary hexadecyl group, a stearyl group, a 2-ethylhexyl group, a 2-butyloctyl group, a 2-butyldecyl group, and a 2-hexyloctyl group.

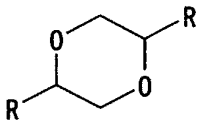
[0018] The alkyl groups preferably have 4-9 carbon atoms, more preferably 4-7 carbon atoms, still more preferably 4-6 carbon atoms, and most preferably 5 carbon atoms.

[0019] While an alkanediol composition according to the present invention may be produced by any process, the alkanediol composition may be produced by oxidizing an olefin into an epoxy compound and subsequently hydrolyzing the epoxy compound. The olefin may be oxidized with an oxidizing agent, such as hydrogen peroxide, a peroxide compound, or an inorganic oxidizing agent. The epoxy compound may be hydrolyzed by a direct reaction with water under high pressure or by a reaction between the epoxy compound and formic acid or acetic acid, followed by hydrolysis with an alkaline substance. Alkanediol compositions thus produced always contain by-products, such as ester compounds, unsaturated alcohols, or acetal compounds. However, compounds causing smells have not been identified. Thus, alkanediol compositions are usually purified by distillation, which is a common purification method, as described above. However, smells of the alkanediol compositions cannot be removed by distillation alone.

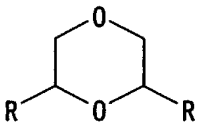
[0020] As a result of investigations, the present inventors found that ester compounds and dioxane compounds have the largest influence on the smell among compounds causing smells. In an alkanediol composition according to a first embodiment of the present invention, therefore, the content of an ester compound (A) must be 0.005 parts by mass or less per 100 parts by mass of alkanediol compound. The term "ester compound (A)" means a compound having an ester bond and includes reaction products between epoxy compounds and fatty acids, such as formic acid and acetic acid, reaction products between alkanediol compounds and fatty acids, and reaction products between compounds having a carbonyl group, which are produced by the oxidation of olefins, and epoxy compounds or alkanediol compounds. These

ester compounds cause a smell. Hence, the content of an ester compound in an alkanediol composition according to the present invention is 0.005 parts by mass or less, preferably 0.003 parts by mass or less, and more preferably 0.001 parts by mass or less per 100 parts by mass of alkanediol compound. More than 0.005 parts by mass of ester compound (A) in an alkanediol composition causes an unpleasant smell.

[0021] In an alkanediol composition according to a second embodiment of the present invention, the content of a dioxane compound (C) must be 0.2 parts by mass or less per 100 parts by mass of alkanediol compound. In the present invention, reduction in the amount of dioxane compound (C) can also reduce a smell. Examples of the dioxane compound (C) include compounds having general formulae (2) and (3) (wherein R is as defined above). The dioxane compound (C) may be produced by a dehydration condensation reaction of alkanediols or a condensation reaction of unreacted epoxy compounds. The content of the dioxane compound (C) in an alkanediol composition according to a second embodiment of the present invention is 0.2 parts by mass or less, preferably 0.15 parts by mass or less, and more preferably 0.1 parts by mass or less per 100 parts by mass of alkanediol compound. More than 0.2 parts by mass of dioxane compound (C) in an alkanediol composition causes an unpleasant smell.

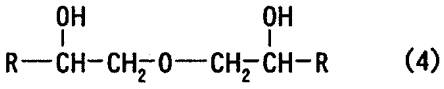


(2)

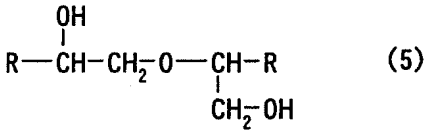


(3)

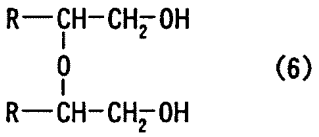
[0022] In the present invention, as described above, reduction of the content of either the ester compound (A) or the dioxane compound (C) leads to a less malodorous alkanediol composition. Another by-product causing a smell of an alkanediol composition is an ether-containing dihydric alcohol. Examples of the ether-containing dihydric alcohol (B) include compounds having general formulae (4), (5), and (6) (wherein R is as defined above). The ether-containing dihydric alcohol (B) may be produced by a dehydration condensation reaction of alkanediols. The content of the ether-containing dihydric alcohol (B) in an alkanediol composition according to the present invention is preferably 0.3 parts by mass or less, more preferably 0.2 parts by mass or less, and still more preferably 0.15 parts by mass or less per 100 parts by mass of alkanediol compound. More than 0.3 parts by mass of ether-containing dihydric alcohol (B) in an alkanediol composition causes an unpleasant smell.



(4)



(5)



(6)

[0023] Such a by-product may be analyzed by a method using an analyzer, such as gas chromatography, liquid chromatography, a mass spectrometer, NMR, or IR, or by titration analysis of a saponification value, the degree of unsaturation, or a hydroxyl value. In terms of microanalysis of a by-product, the by-product may be analyzed preferably by instrumental analysis, more preferably by gas chromatography or with a mass spectrometer, and still more preferably by gas chromatography. In an alkanediol composition according to the present invention, because the peak area ratio in a gas chromatography chart indicates the mass ratio, the mass ratio of each component can easily be determined.

[0024] Gas chromatography analysis may be performed under any condition under which a by-product can be separated. Exemplary conditions are as follows: a gas chromatograph GC-14B (Shimadzu Corporation) with a J & W Scientific DB-1 column (100% dimethylpolysiloxane, inner diameter 0.53 mm, length 15 m, film thickness 1.5 μm); a 20 ml/min nitrogen carrier gas; an FID detector temperature of 320°C; an air pressure of 50 kPa; a hydrogen pressure of 50 kPa; and a vaporization chamber temperature of 320°C. The column may be held at 60°C for five minutes, be heated to 280°C at 10°C/min, and be held at 280°C for three minutes. 0.1 μl of sample diluted to 1.5% by weight in ethanol may be injected with a microsyringe.

[0025] Under the conditions described above, a main peak of 1,2-octanediol appears at a retention time of about 10 min. A peak of a by-product, if any, appears before or after the main peak. The peak area ratio indicates the mass ratio of each component. A peak of an ester compound appears zero to two minutes before the retention time of the main peak. A peak of a dioxane compound appears four to seven minutes after the retention time of the main peak. A peak of an ether-containing dihydric alcohol appears eight to fifteen minutes after the retention time of the main peak.

[0026] In an alkanediol composition according to a first embodiment of the present invention, the peak area of an ester compound appearing zero to two minutes before the retention time of the main peak must be 0.005% or less, preferably 0.003% or less, and more preferably 0.001% or less of the main peak area. When the peak area of an ester compound exceeds 0.005% of the main peak area, the content of the ester compound exceeds 0.005 parts by mass per 100 parts by mass of alkanediol compound. A component causing a smell and having an unknown structure may appear zero to two minutes after the retention time of the main peak. A peak in this area is therefore indicative of a strong smell and is not preferred.

[0027] In an alkanediol composition according to a second embodiment of the present invention, the peak area of a dioxane compound appearing four to seven minutes after the retention time of the main peak must be 0.2% or less, preferably 0.15% or less, and more preferably 0.1% or less of the main peak area. When the peak area of a dioxane compound exceeds 0.2% of the main peak area, the content of the dioxane compound exceeds 0.2 parts by mass per 100 parts by mass of alkanediol compound.

[0028] The peak area of an ether-containing dihydric alcohol appearing eight to fifteen minutes after the retention time of the main peak is preferably 0.3% or less, more preferably 0.2% or less, and still more preferably 0.15% or less of the peak area of the main peak. When the peak area of an ether-containing dihydric alcohol exceeds 0.3% of the main peak area, the content of the ether-containing dihydric alcohol exceeds 0.3 parts by mass per 100 parts by mass of alkanediol compound. This may cause a smell.

[0029] An alkanediol composition according to the present invention may be produced by any known method that can reduce an ester compound by-product to 0.005 parts by mass or less, and/or that can reduce a dioxane compound by-product to 0.2 parts by mass or less, per 100 parts by mass of alkanediol compound. As described above, the content of ester compound and/or the content of dioxane compound cannot be reduced to the range described above by common distillation alone. A smell therefore cannot be removed. This is probably because the boiling points of the components causing a smell, that is, an ester compound and a dioxane compound, are very close to the boiling point of the alkanediol compound.

[0030] Examples of a purification method used in the production of an alkanediol composition according to the present invention include azeotropic distillation of by-products and a large amount of water under reduced pressure; azeotropic distillation of by-products and an organic solvent under reduced pressure; azeotropic distillation of by-products, water, and an organic solvent under reduced pressure; blowing of water vapor under reduced pressure; adsorption of by-products on an adsorbent, such as an ion exchange resin or activated carbon, and subsequent distillation; recrystallization in a solvent, such as ethanol, acetone, or hexane; and combinations thereof; and combinations of these methods and known purification methods.

[0031] Among others, blowing of water vapor under reduced pressure; azeotropic distillation of by-products and water

and/or an organic solvent under reduced pressure (this is a method according to the present invention); and the addition of an organic solvent and blowing of water vapor under reduced pressure are preferred, because an ester compound causing a smell can efficiently be removed by these methods.

[0032] In the blowing of water vapor, the total amount of water is preferably in the range of 30 to 200 parts by mass, more preferably in the range of 50 to 150 parts by mass, and still more preferably in the range of 50 to 100 parts by mass per 100 parts by mass of alkanediol composition. The distillation temperature is preferably in the range of 80°C to 120°C, more preferably in the range of 85°C to 110°C, and still more preferably in the range of 90°C to 100°C. The degree of vacuum is preferably 10 kPa or less, more preferably 5 kPa or less, and still more preferably 1.3 kPa or less.

[0033] The amount of the organic solvent is preferably in the range of 30 to 200 parts by mass, more preferably in the range of 50 to 150 parts by mass, and still more preferably in the range of 80 to 120 parts by mass per 100 parts by mass of alkanediol composition. Furthermore, the organic solvent may be added in a plurality of portions. For example, the addition and removal of 10 parts by mass of organic solvent may be repeatedly performed. In the addition of the organic solvent in a plurality of portions, the total amount of the organic solvent is preferably in the range as described above.

[0034] The amount of water is preferably in the range of 30 to 200 parts by mass, more preferably in the range of 50 to 150 parts by mass, and still more preferably in the range of 80 to 120 parts by mass per 100 parts by mass of alkanediol composition. Furthermore, water may be added in a plurality of portions. For example, the addition and removal of 10 parts by mass of water may be repeatedly performed. In the addition of water in a plurality of portions, the total amount of water is preferably in the range as described above. The degree of vacuum is preferably 10 kPa or less, more preferably 5 kPa or less, and still more preferably 1.3 kPa or less.

[0035] When an organic solvent and water, or an organic solvent and water vapor, are combined, preferably 20-150 parts by mass, more preferably 30-100 parts by mass of water or water vapor per 100 parts by mass of organic solvent is added or blown. The total amount of the organic solvent and water or the total amount of the organic solvent and water vapor is preferably in the range of 50 to 300 parts by mass, more preferably in the range of 80 to 250 parts by mass, and still more preferably in the range of 100 to 200 parts by mass per 100 parts by mass of alkanediol composition. The degree of vacuum is preferably 10 kPa or less, more preferably 5 kPa or less, and still more preferably 1.3 kPa or less.

[0036] Examples of organic solvent for use in the purification method include glycol compounds, such as ethylene glycol, propylene glycol, butylene glycol, diethylene glycol, dipropylene glycol, methyl cellosolve, ethyl cellosolve, propyl cellosolve, butyl cellosolve, propylene glycol monomethyl ether, and diethylene glycol monomethyl ether; acyclic hydrocarbons, such as hexane, octane, nonane, and decane; and aromatic hydrocarbons, such as benzene, toluene, xylene, trimethylbenzene, cumene, and naphthalene. Among others, in terms of removal of by-products, glycol solvents are preferred, propylene glycol, dipropylene glycol, and diethylene glycol are more preferred, and propylene glycol is most preferred.

[0037] If necessary, another component may be added to an alkanediol composition according to the present invention within the limits not compromising the advantages of the present invention. Examples of the additive component include water; surfactants, such as nonionic surfactants, anionic surfactants, cationic surfactants, and amphoteric surfactants; antioxidants; light stabilizers; silicone oils; thickeners; oil solutions; powders (pigments, dyes, and resins); preservatives; perfumes; humectants; bioactive components; salts; solvents; antioxidants; chelating agents; counteractives; pH adjusters; enzymes; and cyclic oligosaccharides such as cyclodextrins.

[0038] An alkanediol composition according to the present invention may be allowed to react with another compound reactive with a hydroxyl group. Examples of the compound reactive with a hydroxyl group include isocyanate compounds, epoxy compounds, and compounds containing carboxylic acids such as fatty acids. An alkanediol composition according to the present invention may be allowed to react with these compounds and thereby be included in urethane resins, epoxy resins, and polyesters. Furthermore, because an alkanediol composition according to the present invention contains only a small amount of by-products, few side reactions occur, and a final product advantageously contains only a small amount of by-products.

[0039] A cosmetic according to the present invention contains an alkanediol composition according to the present invention. In the cosmetic, the alkanediol composition may be used as a humectant or a preservative, or a pearling agent in a shampoo. Examples of the cosmetic include facial cleansing creams, cleansing foams, cleansing creams, cleansing milks, cleansing lotions, massage creams, cold creams, moisture creams, sun blocks, body shampoos, hair shampoos, hair rinses, hair treatments, hair-restorers, hair creams, hair lacquers, setting lotions, hair bleaches, color rinses, permanent wave lotions, hand creams, lipsticks, packs, foundations, toilet water, cosmetic liquid, milky lotions, colognes, bath cosmetics, kitchen detergents, laundry detergents, bath detergents, nail cosmetics, and tooth powders.

EXAMPLES

[0040] The present invention will be further illustrated with examples below.

Production of diol compound

Production Example 1 (Production of octanediol)

[0041] 381 g (3.4 mol) of α-olefin having eight carbon atoms and 218 g (4.5 mol) of 95% formic acid were charged into a 2000 ml four-neck glass flask with a stirrer, a thermometer, and a nitrogen inlet. Then, 261 g (4.6 mol) of 60% hydrogen peroxide was added dropwise at 60°C for two hours. After the completion of dropping, the reactant was aged at 60°C for eight hours. After the reactant was left to stand and a lower aqueous layer was removed, 192 g (1.2 mol) of 25% aqueous sodium hydroxide was added to the reactant. The reactant was stirred at 60°C for 30 minutes for saponification and was left to stand. A lower aqueous layer was then removed. After the saponification and removal of the aqueous layer were performed one more time, the product was washed with hot water and hexane to yield 380 g of 1,2-octanediol.

Production Example 2 (Production of decanediol)

[0042] As in Production Example 1, 1,2-decanediol was produced from an α-olefin having 10 carbon atoms.

Production Example 3 (Production of dodecanediol)

[0043] As in Production Example 1, 1,2-dodecanediol was produced from an α-olefin having 12 carbon atoms. Purification

Treatment 1

[0044] A hundred grams of diol compound produced in Production Example 1, 2, or 3 and 100 g of propylene glycol were charged into a 500 ml four-neck glass flask with a stirrer, a thermometer, and a nitrogen inlet. The reactant was heated to 90°C to 95°C. Propylene glycol was then completely removed at a reduced pressure of 1.33 kPa to yield a purified diol compound.

Treatment 2

[0045] A hundred grams of diol compound produced in Production Example 1, 2, or 3, 100 g of propylene glycol, and 50 g of water were charged into a 500 ml four-neck glass flask with a stirrer, a thermometer, and a nitrogen inlet. The reactant was heated to 90°C to 95°C. Propylene glycol was then completely removed at a reduced pressure of 1.33 kPa to yield a purified diol compound.

Treatment 3

[0046] A hundred grams of diol compound produced in Production Example 1, 2, or 3 was charged into a 300 ml four-neck glass flask with a stirrer, a thermometer, and a nitrogen inlet. The reactant was distilled at a temperature in the range of 120°C to 180°C and a reduced pressure in the range of 1.4 to 1.8 kPa to yield a purified diol compound as a main fraction. Treatment 4

[0047] Fifty grams of diol compound produced in Production Example 1, 2, or 3 and 10 g of activated carbon were charged into a 200 ml beaker and were stirred at 50°C. After two hours, the activated carbon was filtered out to yield a purified diol compound.

[0048] The purified diol compounds thus produced were analyzed by gas chromatography under conditions described below. Peak areas were calculated to determine the amounts of impurities. The amounts of impurities were expressed as ratios of peak area to that of the main peak (diol compound), which was set to be 100. Tables 1 to 3 show the results for 1,2-octanediol (Production Example 1), 1,2-decanediol (Production Example 2), and 1,2-dodecanediol (Production Example 3), respectively.

Analyzer: GC-14B (Shimadzu Corporation)
Column: DB-1 (J & W Scientific, 100% dimethylpolysiloxane, inner diameter 0.53 mm, length 15 m, film thickness 1.5 μm)
Carrier gas: Nitrogen, 20 ml/min
Air pressure: 50 kPa
Hydrogen pressure: 50 kPa
Temperature of vaporization chamber: 320°C
Heating rate: held at 60°C for five minutes, heated to 280°C at 10°C/min, and held at 280°C for three minutes.

[0049] Sample amount: 0.1 μl (1.5% by weight in ethanol)

Table 1

	Retention time (min)				
	8.3~10.2	10.3	10.4~12.3	14.3~17.3	18.3~25.3
Treatment 1	0.003	100	0	0.16	0.29
Treatment 2	0.001	100	0	0.03	0.25
Treatment 3	0.03	100	0	0.29	0.41
Treatment 4	0.13	100	0	0.32	0.48
Untreated	0.25	100	0.05	0.48	0.58
Identified components	Ester compound	Diol compound	-	Dioxane compound	Ether-containing dihydric alcohol

Table 2

	Retention time (min)				
	11.1~13.0	13.1	13.2~15.1	17.1~20.1	21.1~28.1
Treatment 1	0.006	100	0	0.18	0.32
Treatment 2	0.002	100	0	0.04	0.23
Treatment 3	0.05	100	0	0.30	0.42
Treatment 4	0.14	100	0	0.33	0.51
Untreated	0.23	100	0.04	0.46	0.61
Identified components	Ester compound	Diol compound	-	Dioxane compound	Ether-containing dihydric alcohol

Table 3

	Retention time (min)				
	13.7~15.6	15.7	15.8~17.7	19.7~22.7	23.7~30.7
Treatment 1	0.006	100	0	0.19	0.29
Treatment 2	0.002	100	0	0.06	0.21
Treatment 3	0.05	100	0	0.25	0.41
Treatment 4	0.15	100	0	0.32	0.49
Untreated	0.21	100	0.03	0.45	0.6
Identified components	Ester compound	Diol compound	-	Dioxane compound	Ether-containing dihydric alcohol

Test Example 1

[0050] Alkanediol compounds in Treatments 1 to 4 and untreated alkanediol compounds were evaluated by ten panelists (five men and five women) for evaluation items described below. Table 4 shows the results.

O: No smell.
Δ: A little smell but not unpleasant.
×: Unpleasant smell.

Table 4

		Number of Panelists		
		○	△	×
Treatment 1	1,2-octanediol	9	1	0
	1,2-decanediol	8	2	0
	1,2-dodecanediol	9	1	0
Treatment 2	1,2-octanediol	10	0	0
	1,2-decanediol	10	0	0
	1,2-dodecanediol	10	0	0
Treatment 3	1,2-octanediol	0	4	6
	1,2-decanediol	0	3	7
	1,2-dodecanediol	0	4	6
Treatment 4	1,2-octanediol	0	2	8
	1,2-decanediol	0	1	9
	1,2-dodecanediol	0	1	9
Untreated	1,2-octanediol	0	0	10
	1,2-decanediol	0	0	10
	1,2-dodecanediol	0	0	10

[0051] These results show that, in gas chromatography, a large peak (ester compound) at zero to two minutes before the peak of a diol compound and/or a large peak (dioxane compound) at four to seven minutes after the peak of the diol compound are indicative of an unpleasant smell.

Test Example 2

[0052] 1,2-octanediol in Production Example 1 was purified by the method described in Treatment 2. Shampoos were prepared with the purified 1,2-octanediol and untreated 1,2-octanediol according to the formula shown in Table 5. The two shampoos were evaluated by ten panelists (five men and five women) for smell when the shampoos were used.

Table 5

Component	Amount (% by mass)
Sodium polyoxyethylene (3) lauryl ether sulfate	25.0
Lauryl amine oxide	3.0
Lauryl amine propyl betaine	18.0
Propylene glycol	3.2
Hydroxyethyl cellulose	0.5

(continued)

Component	Amount (% by mass)
1,2-octanediol	0.1
Purified water	Remainder

[0053] As a result of evaluating the smell of shampoo, nine panelists felt that the smell of the shampoo containing a 1,2-octanediol composition purified by the method described in Treatment 2 was better than that of the shampoo containing an untreated 1,2-octanediol composition; one panelist felt that the two shampoos have the same smell; and no panelist felt that the smell of the shampoo containing an untreated 1,2-octanediol composition was better than that of the shampoo containing a purified 1,2-octanediol composition.

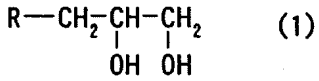
Claims

1. An alkanediol composition comprising 0.005 parts by mass or less of ester compound (A) per 100 parts by mass of alkanediol compound having four or more carbon atoms.

2. An alkanediol composition comprising 0.2 parts by mass or less of dioxane compound (C) per 100 parts by mass of alkanediol compound having four or more carbon atoms.

3. The alkanediol composition according to Claim 1 or 2, further comprising 0.3 parts by mass or less of ether-containing dihydric alcohol (B) per 100 parts by mass of the alkanediol compound.

4. The alkanediol composition according to any one of Claims 1 to 3, wherein the alkanediol compound has a general formula (1):



(1)

wherein R denotes an alkyl group having 4-15 carbon atoms.

5. The alkanediol composition according to any one of Claims 1 to 4, wherein the alkanediol compound is a hydrolysate of an epoxy compound produced by an oxidation reaction of an olefin.

6. A process for producing an alkanediol composition, comprising the steps of:

   (a) oxidizing an olefin to produce an epoxy compound;
   (b) hydrolyzing the epoxy compound to produce the alkanediol composition; and
   (c) adding water and/or an organic solvent to the alkanediol composition and removing water and/or the organic solvent under reduced pressure.

7. A cosmetic comprising the alkanediol composition according to any one of Claims 1 to 5.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2006/308652

A. CLASSIFICATION OF SUBJECT MATTER
C07C29/10(2006.01), ***A61K8/30***(2006.01), ***C07C29/80***(2006.01), ***C07C31/20***(2006.01)

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C07C29/10(2006.01), ***A61K8/30***(2006.01), ***C07C29/80***(2006.01), ***C07C31/20***(2006.01)

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
Jitsuyo Shinan Koho 1922-1996 Jitsuyo Shinan Toroku Koho 1996-2006
Kokai Jitsuyo Shinan Koho 1971-2006 Toroku Jitsuyo Shinan Koho 1994-2006

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X A	JP 2000-505103 A (LG Chemical Ltd.), 25 April, 2000 (25.04.00), Table 6 & US 6201159 B1 & WO 98/017614 A1	2-3 1
X Y A	JP 61-65834 A (Daicel Chemical Industries, Ltd.), 04 April, 1986 (04.04.86), Claims; page 1, lower right column, lines 3 to 9; example 1 (Family: none)	2-3,7 6 1
Y A	JP 7-10789 A (Daicel Chemical Industries, Ltd.), 13 January, 1995 (13.01.95), Claim 1 & US 5345004 A & EP 616992 A1	6 1

☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" earlier application or patent but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
17 May, 2006 (17.05.06)	30 May, 2006 (30.05.06)
Name and mailing address of the ISA/ Japanese Patent Office Facsimile No.	Authorized officer Telephone No.

Form PCT/ISA/210 (second sheet) (April 2005)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2006/308652

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y A	JP 61-130247 A (Degussa AG.), 18 June, 1986 (18.06.86), Claim 1 & US 4605795 A & EP 183029 A2 & DE 3442937 A1	6 5
A	JP 2004-115404 A (Kyowa Yuka Co., Ltd.), 15 April, 2004 (15.04.04), Par. Nos. [0009], [0019] (Family: none)	4,7

Form PCT/ISA/210 (continuation of second sheet) (April 2005)

REFERENCES CITED IN THE DESCRIPTION

This list of references cited by the applicant is for the reader's convenience only. It does not form part of the European patent document. Even though great care has been taken in compiling the references, errors or omissions cannot be excluded and the EPO disclaims all liability in this regard.

Patent documents cited in the description

- JP 57062234 A **[0005]**
- JP 60078928 A **[0006]**

Third Party Submission

U.S. Serial No. 16/262,902 (US2019/0289848)

Cite No.	Reference	Concise Description of Relevance
1	De Bruyn et al., US20050065389 (March 24, 2005)	De Bruyn et al. describe alcohols formed from starting olefinic compounds, thus covering 1,2-alkanediols according to pending claim 42 of US2019/0289848. De Bruyn et al. provide a process of increasing the carbon chain length of an olefinic compound comprising the steps of providing a starting olefinic compound and subjecting it to hydroformylation to produce an aldehyde and/or alcohol with an increased carbon chain length compared to the starting olefinic compound. Optionally the aldehyde that may form during the hydroformylation reaction is hydrogenated to convert it to an alcohol which has an increased carbon chain length compared to the starting olefinic compound. The alcohol with the increased carbon chain length is subjected to dehydration to produce an olefinic compound with an increased carbon chain length compared to the starting olefinic compound. See abstract. In one embodiment of the invention a Fischer-Tropsch derived feed stream containing one or more α-olefins may be used as a source of the starting olefinic compound. See paragraph [0017]. It is common knowledge that Fischer-Tropsch synthesis allows production of liquid hydrocarbons fuel from biomass. Accordingly, the alcohols formed from starting olefinic compounds according to embodiments of De Bruyn et al. are non-petrochemically-derived, i.e., they cover "bio"- 1,2-alkanediols according to pending claim 42 of US2019/0289848. See paragraph [0012] of US2019/0289848.
2	Wu et al., US4234752 (November 18, 1980)	Wu et al. describe the conversion of alcohols to alkenes, which alkenes can serve as starting materials for forming 1,2-alkanediols according to pending claim 42 of US2019/0289848. In an embodiment, Wu et al. describe the production of 3-Methyl-1-butene by dehydrating 3-Methyl-1-butanol. See abstract.
3	Fuji et al., US20030065233 (April 3, 2003)	Fuji et al. describe a production process for α-olefins making use of elimination reaction of a primary alcohol or an ether (see Abstract), which α-olefins can serve as starting materials for forming bio-1,2-alkanediols according to pending claim 42 of US2019/0289848. In the process, unreacted material and ether intermediate byproduct produced in the process can be used in the reaction with the same catalyst. This allows industrially and environmentally favorable production process of α-olefins. See abstract.
4	Swern et al., *Hydroxylation and Epoxidation of Some 1-Olefins with Per-acids*, JACS, 68:1504-1507 (1946)	Swern et al. describe 1,2-alkanediols according to pending claim 42 of US2019/0289848. The 1,2-alkanediols, called 1,2-glycols, were prepared from straight-chain 1-olefins. 1,2-octanediol, 1,2-decanediol, 1,2-dodecanediol and 1,2-tetradecanediol were prepared for the first time. See Summary.
5	Osterholt et al., US2006161025 (June 10, 2008)	Osterholt et al. describe 1,2-alkanediols according to pending claim 42 of US2019/0289848. E.g. Osterholt et al. describe the production of 1,2-octanediol from 1-octene. See Ex. 5, para. [0091]-[0096].
6	Furuya et al., EP1876162A1 (January 9, 2008)	Furaya et al. describe an alkanediol composition containing 0.005 parts by mass or less of ester compound per 100 parts by mass of alkanediol compound having four or more carbon atoms, and an alkanediol composition contains 0.2 parts by mass or less of dioxane compound per 100 parts by mass of alkanediol compound having four or more carbon atoms. See Abstract. Accordingly, Furaya is of relevance for "bio"-1,2-alkanediols according to pending claim 42 of US2019/0289848.

Electronic Patent Application Fee Transmittal

Application Number:	16262902
Filing Date:	
Title of Invention:	
First Named Inventor/Applicant Name:	
Filer:	Daniel Altman
Attorney Docket Number:	

Filed as Large Entity

Filing Fees for

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
DOCUMENT FEE FOR THIRD-PARTY SUBMISSIONS	1818	1	180	180
Pages:				
Claims:				
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Extension-of-Time:				
Miscellaneous:				
			Total in USD ($)	**180**

Electronic Acknowledgement Receipt

EFS ID:	39458564
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	
First Named Inventor/Applicant Name:	
Correspondence Address:	- - - - - - - - - -
Filer:	Daniel Altman
Filer Authorized By:	
Attorney Docket Number:	
Receipt Date:	15-MAY-2020
Filing Date:	
Time Stamp:	18:42:45
Application Type:	

Payment information:

Submitted with Payment	yes
Payment Type	CARD
Payment was successfully received in RAM	$180

RAM confirmation Number	E20205EI42433792
Deposit Account	
Authorized User	

The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows:

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Concise Description of Relevance	Concise-description-generated.pdf	31750 615865e64d82dfadc6351aa94a0632d64401ef7b	no	3
Warnings:					
Information:					
2	Third-Party Submission Under 37 CFR 1.290	Third-party-preissuance-submission.pdf	46741 002e68dcd3a0f50630d9d4ebe17ced96d2dc5669	no	3
Warnings:					
Information:					
3	Request for Notification of Non-compliant Third-Party Submission	Third-party-notification-request.pdf	19634 5de84c2d0014c0feb2351d49582dd59a8607e04a	no	1
Warnings:					
Information:					
4	Non Patent Literature	Swern_1946.pdf	6800133 d53ccafeefc62196697c8c077714ed9709cfd112	no	5
Warnings:					
Information:					
5	Foreign Reference	EP1876162A1.pdf	235184 5791cf9dd1af65c6954d610eb43be963606cd9b8	no	13
Warnings:					
Information:					

6	Concise Description of Relevance	AttachedCociseExplanations.pdf	130271 8b8bb7f08059692cef8e377cb321bccf320dd822	no	1
Warnings:					
Information:					
7	Fee Worksheet (SB06)	fee-info.pdf	29140 cc5b72ff717f80f7302274dd0f42be2420f72661	no	2
Warnings:					
Information:					
		Total Files Size (in bytes):	7292853		

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	I0113-198U2	1028

159443 7590 05/19/2020

Calderone Bullock LLC
Two Greentree Centre, Suite 301
9000 Lincoln Drive East
Marlton, NJ 08053

EXAMINER	
HOLLOMAN, NANNETTE	

ART UNIT	PAPER NUMBER
1612	

NOTIFICATION DATE	DELIVERY MODE
05/19/2020	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

USP@calderonebullock.com
docketing@calderonebullock.com
kbullock@calderonebullock.com

UNITED STATES DEPARTMENT OF COMMERCE
U.S. Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450

APPLICATION NO./ CONTROL NO.	FILING DATE	FIRST NAMED INVENTOR/ PATENT IN REEXAMINATION	ATTORNEY DOCKET NO.
16/262,902	01/30/2019	Burgo et al.	I0113-198U2

		EXAMINER	
Calderone Bullock LLC Two Greentree Centre, Suite 301 9000 Lincoln Drive East Marlton, NJ 08053		MARIANNE C SEIDEL	
		ART UNIT	PAPER
		1600	20200518

DATE MAILED: _____

Please find below and/or attached an Office communication concerning this application or proceeding.

Commissioner for Patents

The third-party submission under 37 CFR 1.290 filed on 5/15/2020 for the instant application has been determined to be compliant with 35 U.S.C. 122(e) and 37 CFR 1.290 and is being entered in the application. Please allow a few days for the submission to be visible in the Patent Application Information Retrieval (PAIR) system.

/MARIANNE C SEIDEL/
Quality Assurance Specialist, Art Unit 1600

PTO-90C (Rev.04-03)

Third Party Submission

U.S. Serial No. 16/262,902 (US2019/0289848)

Cite No.	Reference	Concise Description of Relevance
1	Fortman et al., US9856461B2 (March 10, 2016)	Fortman et al. describe a polyketide synthase (PKS) capable of synthesizing an α-olefin, such as 1-hexene or butadiene. See summary. Fortman et al. specifically disclose 1-alkenes such as 1-hexene, 1-pentene, 1-butene, and propylene (column 14, lines 49-52). Precursor molecules are said to be biosynthesized (column 22, lines 12-18), so synthesized α-olefins can also be interpreted to be non-petrochemically-derived, i.e. they cover "bio"-1-alkenes according to pending claim 42 of US2019/0289848. See paragraph [0012] of US2019/0289848. The "bio"-α-olefins according to Fortman et al. can serve as starting materials for forming "bio"-1,2-alkanediols according to pending claim 42 of US2019/0289848.
2	Klug et al., *Degradation of Hydrocarbons by Members of the Genus Candida, II. Oxidation of n-Alkanes and 1-Alkenes by Candida lipolytica*, Journal of Bacteriology, Vol. 93, No. 6, pp 1847-1852 (1967)	Klug et al. describe Pseudomonas-mediated epoxidation of an 1-octene double bond followed by nonenzymatic rupture of the epoxide bond to yield 1,2-octanediol (page 1851, right column, lines 14-28). Klug et al. do not describe that the 1-octene is petrochemically-derived. Klug et al. are thus considered to disclose a "bio"-1,2-alkanediol formed by conversion of a "bio"-1-alkene, according to pending claim 42 of US2019/0289848.
3	Liu et al., *Hydrogen peroxide-independent production of α-alkenes by OleT$_{JE}$ P450 fatty acid decarboxylase*, Biotechnology for Biofuels, 7:28 (2014)	Liu et al. describe a Jeotgalicoccus sp.-mediated decarboxylation of C12-C20 fatty acids to alpha-olefins. See Results section of Abstract. Also it is described that the alpha-olefines are biogenic α-alkenes, which are renewable and more environmentally friendly than petroleum-derived equivalents. See Background section of Abstract. Liu et al. thus describe "bio"-α-alkenes that can serve as starting materials for forming "bio"-1,2-alkanediols according to pending claim 42 of US2019/0289848.
4	Wright, US8912373B2 (September 20, 2012)	Wright describes the dehydration of bio-1-alcohols to bio-1-alkenes with high selectivity. See column 3, line 54-column 4, line 26. Fig. 1 illustrates a general overview of creating renewable and sustainable biodiesel, biojet, and biolubricants based on a feed stream of bio-1-alcohols. It is clear that this are non-petrochemically-derived products, as is evidenced by the starting materials, e.g. cellulosic and hemi-cellulosic biomass, in Fig. 1. Wright thus covers "bio"-1-alkenes according to pending claim 42 of US2019/0289848. The "bio"-1-alkenes according to wright can serve as starting materials for forming "bio"-1,2-alkanediols according to pending claim 42 of US2019/0289848.

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290
CONCISE DESCRIPTION OF RELEVANCE

Application Number	16262902

U.S. PATENTS

Cite No	Patent Number	Concise Description of Relevance
1	9856461	See attached
2	8912373	See attached

U.S. PATENT APPLICATION PUBLICATION

Cite No	Publication Number	Concise Description of Relevance

FOREIGN PATENT DOCUMENTS		
CiteNo	**Foreign Document Number**	**Concise Description of Relevance**

NON-PATENT PUBLICATIONS		
Cite No	**Reference**	**Concise Description of Relevance**

1	Liu et al., Hydrogen peroxide-independent production of α-alkenes by OleTJE P450 fatty acid decarboxylase, Biotechnology for Biofuels, 7:28 (2014)	See attached

2	Klug et al., Degradation of Hydrocarbons by Members of the Genus Candida, II. Oxidation of n-Alkanes and 1-Alkenes by Candida lipolytica, Journal of Bacteriology, Vol. 93, No. 6, pp 1847-1852 (1967)	See attached

PTO/SB/429(08-12)
Approved for use through 07/31/2015. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

U.S. PATENTS

Cite No	Patent Number	Kind Code[1]	Issue Date (YYYY-MM-DD)	First Named Inventor
1	8912373	B2	2014-12-16	Michael Wright
2	9856461	B2	2018-01-02	Jeffrey Fortman

U.S. PATENT APPLICATION PUBLICATIONS

Cite No	Publication Number	Kind Code[1]	Publication Date (YYYY-MM-DD)	First Named Inventor

FOREIGN PATENTS AND PUBLISHED FOREIGN PATENT APPLICATIONS

Cite No	Foreign Document Number[3]	Country Code[2]	Kind Code[1]	Publication Date (YYYY-MM-DD)	Applicant, Patentee or First Named Inventor	T[5]
						☐

NON-PATENT PUBLICATIONS (e.g., journal article, Office action)

Cite No	Author (if any), title of the publication, page(s) being submitted, publication date, publisher (where available), place of publication (where available).	T[5]	E[6]

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

1	Klug et al., Degradation of Hydrocarbons by Members of the Genus Candida, II. Oxidation of n-Alkanes and 1-Alkenes by Candida lipolytica, Journal of Bacteriology, Vol. 93, No. 6, pp 1847-1852 (1967)	☐	☐
2	Liu et al., Hydrogen peroxide-independent production of α-alkenes by OleTJE P450 fatty acid decarboxylase, Biotechnology for Biofuels, 7:28 (2014)	☐	☐

STATEMENTS

The party making the submission is not an individual who has a duty to disclose information with respect to the above-identified application under 37 CFR 1.56.

This submission complies with the requirements of 35 U.S.C. 122(e) and 37 CFR 1.290.

☒ The fee set forth in 37 CFR 1.290(f) has been submitted herewith.

☐ The fee set forth in 37 CFR 1.290(f) is not required because this submission lists three or fewer total items and, to the knowledge of the person signing the statement after making reasonable inquiry, this submission is the first and the only submission under 35 U.S.C 122(e) filed in the above-identified application by the party making the submission or by a party in privity with the party.

☐ This resubmission is being made responsive to a notification of non-compliance issued for an earlier filed third-party submission. The corrections in this resubmission are limited to addressing the non-compliance. As such, the party making this resubmission: (1) requests that the Office apply the previously-paid fee set forth in 37 CFR 1.290(f), or (2) states that no fee is required to accompany this resubmission as the undersigned is again making the fee exemption statement set forth in 37 CFR 1.290(g).

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

Signature	/daniel altman/		
Name/Print	Daniel Altman	Registration Number (if applicable)	34115

Examiner Signature		Date Considered	

*EXAMINER: Signature indicates all documents listed above have been considered,except for citations through which a line is drawn. Draw line through citation if not considered. Include a copy of this form with next communication to applicant. 1. If known, enter kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16. See MPEP 901.04(a). 2. Enter the country or patent office that issued the document, by two-letter code under WIPO standard ST.3. See MPEP 1851. 3. For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. 4. If known, enter the kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 . See MPEP 901.04(a). 5. Check mark indicates translation attached. 6. Check mark indicates evidence of publication attached.

Electronic Patent Application Fee Transmittal

Application Number:	16262902
Filing Date:	
Title of Invention:	
First Named Inventor/Applicant Name:	
Filer:	Daniel Altman
Attorney Docket Number:	

Filed as Large Entity

Filing Fees for

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
DOCUMENT FEE FOR THIRD-PARTY SUBMISSIONS	1818	1	180	180
Pages:				
Claims:				
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Extension-of-Time:				
Miscellaneous:				
			Total in USD ($)	**180**

Electronic Acknowledgement Receipt

EFS ID:	39582112
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	
First Named Inventor/Applicant Name:	
Correspondence Address:	- - - - - - - - - -
Filer:	Daniel Altman
Filer Authorized By:	
Attorney Docket Number:	
Receipt Date:	29-MAY-2020
Filing Date:	
Time Stamp:	20:26:06
Application Type:	

Payment information:

Submitted with Payment	yes
Payment Type	CARD
Payment was successfully received in RAM	$180

RAM confirmation Number	E20205SK26036637
Deposit Account	
Authorized User	

The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows:

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Concise Description of Relevance	Concise_Statement_of_Relevance.pdf	134079 60aa28d22639c4faaac25636e8c95804038a9123	no	1
Warnings:					
Information:					
2	Non Patent Literature	Liu.pdf	1010390 aa575eb65cdf8baec18ab8176ca614bfa086f423	no	12
Warnings:					
Information:					
3	Non Patent Literature	Klug.pdf	923731 bf2a4ad9c3ad2425039a18b2712a5f5743b2dd92	no	6
Warnings:					
Information:					
4	Concise Description of Relevance	Concise-description-generated.pdf	32941 52f9e254a7c07e8234c593c20682f4d8787e6005	no	4
Warnings:					
Information:					
5	Third-Party Submission Under 37 CFR 1.290	Third-party-preissuance-submission.pdf	45747 15f78eb6e4e8795338a0c4e9de7c226f2e064799	no	3
Warnings:					
Information:					

6	Request for Notification of Non-compliant Third-Party Submission	Third-party-notification-request.pdf	19634 24b173c5b121dfaed9da1f81621571b88cf76993	no	1
Warnings:					
Information:					
7	Fee Worksheet (SB06)	fee-info.pdf	29141 86d3850b1da5df4e31c525965beff6c5181400a9	no	2
Warnings:					
Information:					
		Total Files Size (in bytes):	2195663		

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	I0113-198U2	1028

159443 7590 06/03/2020

Calderone Bullock LLC
Two Greentree Centre, Suite 301
9000 Lincoln Drive East
Marlton, NJ 08053

EXAMINER	
HOLLOMAN, NANNETTE	

ART UNIT	PAPER NUMBER
1612	

NOTIFICATION DATE	DELIVERY MODE
06/03/2020	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

USP@calderonebullock.com
docketing@calderonebullock.com
kbullock@calderonebullock.com

UNITED STATES DEPARTMENT OF COMMERCE
U.S. Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450

APPLICATION NO./ CONTROL NO.	FILING DATE	FIRST NAMED INVENTOR/ PATENT IN REEXAMINATION	ATTORNEY DOCKET NO.
16/262,902	01/30/2019	Burgo et al.	I0113-198U2

		EXAMINER	
Calderone Bullock LLC Two Greentree Centre, Suite 301 9000 Lincoln Drive East Marlton, NJ 08053		MARIANNE C SEIDEL	
		ART UNIT	PAPER
		1600	20200601

DATE MAILED: _____

Please find below and/or attached an Office communication concerning this application or proceeding.

Commissioner for Patents

The third-party submission under 37 CFR 1.290 filed on 5/29/2020 for the instant application has been determined to be compliant with 35 U.S.C. 122(e) and 37 CFR 1.290 and is being entered in the application. Please allow a few days for the submission to be visible in the Patent Application Information Retrieval (PAIR) system.

/MARIANNE C SEIDEL/
Quality Assurance Specialist, Art Unit 1600

PTO-90C (Rev.04-03)

PTO/AIA/82A (07-13)
Approved for use through 03/31/2021. OMB 0651-0035
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

TRANSMITTAL FOR POWER OF ATTORNEY TO ONE OR MORE REGISTERED PRACTITIONERS

NOTE: This form is to be submitted with the Power of Attorney by Applicant form (PTO/AIA/82B) to identify the application to which the Power of Attorney is directed, in accordance with 37 CFR 1.5, unless the application number and filing date are identified in the Power of Attorney by Applicant form. If neither form PTO/AIA/82A nor form PTO/AIA82B identifies the application to which the Power of Attorney is directed, the Power of Attorney will not be recognized in the application.

Application Number	16/262,902
Filing Date	January 30, 2019
First Named Inventor	Rocco Burgo
Title	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
Art Unit	1612
Examiner Name	HOLLOMAN, NANNETTE
Attorney Docket Number	I0113-198U2

SIGNATURE of Applicant or Patent Practitioner

Signature	/Eric J. Choi/	Date (Optional)	
Name	Eric J. Choi	Registration Number	71,160
Title (if Applicant is a juristic entity)	Attorney		
Applicant Name (if Applicant is a juristic entity)	Inolex Investment Corporation		

NOTE: This form must be signed in accordance with 37 CFR 1.33. See 37 CFR 1.4(d) for signature requirements and certifications. If more than one applicant, use multiple forms.

☑ *Total of 1 forms are submitted.

This collection of information is required by 37 CFR 1.131, 1.32, and 1.33. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.11 and 1.14. This collection is estimated to take 3 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

Doc Code: PA..
Document Description: Power of Attorney

PTO/AIA/82B (07-13)
Approved for use through 03/31/2021. OMB 0651-0035
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number

POWER OF ATTORNEY BY APPLICANT

I hereby revoke all previous powers of attorney given in the application identified in either the attached transmittal letter or the boxes below.

Application Number	Filing Date

(Note: The boxes above may be left blank if information is provided on form PTO/AIA/82A.)

☑ I hereby appoint the Patent Practitioner(s) associated with the following Customer Number as my/our attorney(s) or agent(s), and to transact all business in the United States Patent and Trademark Office connected therewith for the application referenced in the attached transmittal letter (form PTO/AIA/82A) or identified above: 171052

OR

☐ I hereby appoint Practitioner(s) named in the attached list (form PTO/AIA/82C) as my/our attorney(s) or agent(s), and to transact all business in the United States Patent and Trademark Office connected therewith for the patent application referenced in the attached transmittal letter (form PTO/AIA/82A) or identified above. (Note: Complete form PTO/AIA/82C.)

Please recognize or change the correspondence address for the application identified in the attached transmittal letter or the boxes above to:

☑ The address associated with the above-mentioned Customer Number

OR

☐ The address associated with Customer Number:

OR

☐ Firm or Individual Name					
Address					
City		State		Zip	
Country					
Telephone		Email			

I am the Applicant (if the Applicant is a juristic entity, list the Applicant name in the box):

INOLEX Investment Corporation

☐ Inventor or Joint Inventor (title not required below)
☐ Legal Representative of a Deceased or Legally Incapacitated Inventor (title not required below)
☑ Assignee or Person to Whom the Inventor is Under an Obligation to Assign (provide signer's title if applicant is a juristic entity)
☐ Person Who Otherwise Shows Sufficient Proprietary Interest (e.g., a petition under 37 CFR 1.46(b)(2) was granted in the application or is concurrently being filed with this document) (provide signer's title if applicant is a juristic entity)

SIGNATURE of Applicant for Patent

The undersigned (whose title is supplied below) is authorized to act on behalf of the applicant (e.g., where the applicant is a juristic entity).

Signature		Date (Optional)	5.22.2020
Name	DAVID C.A. PLIMPTON		
Title	CEO & PRESIDENT		

NOTE: Signature - This form must be signed by the applicant in accordance with 37 CFR 1.33. See 37 CFR 1.4 for signature requirements and certifications. If more than one applicant, use multiple forms.

☐ Total of forms are submitted.

This collection of information is required by 37 CFR 1.131, 1.32, and 1.33. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.11 and 1.14. This collection is estimated to take 3 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

PTO/AIA/82C (07-13)
Approved for use through 03/31/2021. OMB 0651-0035
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number

POWER OF ATTORNEY BY APPLICANT

No more than ten (10) patent practitioners total may be appointed as set forth below by name and registration number. This page need not be submitted if appointing the Patent Practitioner(s) associated with a Customer Number (see form PTO/AIA/82B):

Name	Registration Number

Privacy Act Statement

The **Privacy Act of 1974 (P.L. 93-579)** requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C. 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether the Freedom of Information Act requires disclosure of these records.
2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.
3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.
4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).
5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.
6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).
7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.
8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspections or an issued patent.
9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

Electronic Acknowledgement Receipt

EFS ID:	40277571
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	159443
Filer:	Eric J. Choi/Valerie Dotson
Filer Authorized By:	Eric J. Choi
Attorney Docket Number:	I0113-198U2
Receipt Date:	13-AUG-2020
Filing Date:	30-JAN-2019
Time Stamp:	15:36:41
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Power of Attorney	INLX_0198U2_484799_I0113-198U2_PowerOfAttorney.pdf	331168 cda766b9bad55bcc561fcbb4c479c1bf9640e4c6	no	4

Warnings:

Information:	
Total Files Size (in bytes):	331168

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NUMBER	FILING OR 371(C) DATE	FIRST NAMED APPLICANT	ATTY. DOCKET NO./TITLE
16/262,902	01/30/2019	Rocco V. Burgo	I0113-198U2

CONFIRMATION NO. 1028

POWER OF ATTORNEY NOTICE

159443
Calderone Bullock LLC
Two Greentree Centre, Suite 301
9000 Lincoln Drive East
Marlton, NJ 08053

OC000000119247488

Date Mailed: 08/18/2020

NOTICE REGARDING CHANGE OF POWER OF ATTORNEY

This is in response to the Power of Attorney filed 08/13/2020.

- The Power of Attorney to you in this application has been revoked by the applicant. Future correspondence will be mailed to the new address of record(37 CFR 1.33).

Questions about the contents of this notice and the requirements it sets forth should be directed to the Office of Data Management, Application Assistance Unit, at **(571) 272-4000** or **(571) 272-4200** or **1-888-786-0101**.

/ytdemisse/

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NUMBER	FILING OR 371(C) DATE	FIRST NAMED APPLICANT	ATTY. DOCKET NO./TITLE
16/262,902	01/30/2019	Rocco V. Burgo	I0113-198U2

CONFIRMATION NO. 1028

POA ACCEPTANCE LETTER

171052
Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

OC00000019247520

Date Mailed: 08/18/2020

NOTICE OF ACCEPTANCE OF POWER OF ATTORNEY

This is in response to the Power of Attorney filed 08/13/2020.

The Power of Attorney in this application is accepted. Correspondence in this application will be mailed to the above address as provided by 37 CFR 1.33.

Questions about the contents of this notice and the requirements it sets forth should be directed to the Office of Data Management, Application Assistance Unit, at **(571) 272-4000** or **(571) 272-4200** or **1-888-786-0101**.

/ytdemisse/

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	I0113-198U2	1028

171052 7590 09/17/2020

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

EXAMINER	
HOLLOMAN, NANNETTE	

ART UNIT	PAPER NUMBER
1612	

NOTIFICATION DATE	DELIVERY MODE
09/17/2020	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

echoi@cozen.com
nyipdocketing@cozen.com
vdotson@cozen.com

Office Action Summary	Application No. 16/262,902	Applicant(s) Burgo et al.	
	Examiner NANNETTE HOLLOMAN	Art Unit 1612	AIA (FITF) Status Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTHS FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

1)☑ Responsive to communication(s) filed on 01/30/2019.

☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____.

2a)☐ This action is **FINAL.** 2b) ☑ This action is non-final.

3)☐ An election was made by the applicant in response to a restriction requirement set forth during the interview on _____; the restriction requirement and election have been incorporated into this action.

4)☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims*

5) ☑ Claim(s) 42-80 is/are pending in the application.

5a) Of the above claim(s) _____ is/are withdrawn from consideration.

6) ☐ Claim(s) _____ is/are allowed.

7) ☑ Claim(s) 42-80 is/are rejected.

8) ☐ Claim(s) _____ is/are objected to.

9) ☐ Claim(s) _____ are subject to restriction and/or election requirement

* If any claims have been determined allowable, you may be eligible to benefit from the **Patent Prosecution Highway** program at a participating intellectual property office for the corresponding application. For more information, please see http://www.uspto.gov/patents/init_events/pph/index.jsp or send an inquiry to **PPHfeedback@uspto.gov.**

Application Papers

10)☐ The specification is objected to by the Examiner.

11)☐ The drawing(s) filed on _____ is/are: a)☐ accepted or b)☐ objected to by the Examiner.

Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).

Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).

Priority under 35 U.S.C. § 119

12)☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

Certified copies:

a)☐ All b)☐ Some** c)☐ None of the:

1.☐ Certified copies of the priority documents have been received.

2.☐ Certified copies of the priority documents have been received in Application No. _____.

3.☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

** See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

1) ☑ Notice of References Cited (PTO-892)

2) ☑ Information Disclosure Statement(s) (PTO/SB/08a and/or PTO/SB/08b) Paper No(s)/Mail Date _____.

3) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date _____.

4) ☐ Other: _____.

DETAILED ACTION

Notice of Pre-AIA or AIA Status

The present application, filed on or after March 16, 2013, is being examined under the first inventor to file provisions of the AIA.

Claim Rejections - 35 USC § 103

In the event the determination of the status of the application as subject to AIA 35 U.S.C. 102 and 103 (or as subject to pre-AIA 35 U.S.C. 102 and 103) is incorrect, any correction of the statutory basis for the rejection will not be considered a new ground of rejection if the prior art relied upon, and the rationale supporting the rejection, would be the same under either status.

The following is a quotation of 35 U.S.C. 103 which forms the basis for all obviousness rejections set forth in this Office action:

> A patent for a claimed invention may not be obtained, notwithstanding that the claimed invention is not identically disclosed as set forth in section 102, if the differences between the claimed invention and the prior art are such that the claimed invention as a whole would have been obvious before the effective filing date of the claimed invention to a person having ordinary skill in the art to which the claimed invention pertains. Patentability shall not be negated by the manner in which the invention was made.

The factual inquiries for establishing a background for determining obviousness under 35 U.S.C. 103 are summarized as follows:

1. Determining the scope and contents of the prior art.

2. Ascertaining the differences between the prior art and the claims at issue.

3. Resolving the level of ordinary skill in the pertinent art.

4. Considering objective evidence present in the application indicating obviousness or nonobviousness.

This application currently names joint inventors. In considering patentability of the claims the examiner presumes that the subject matter of the various claims was commonly owned as of the

effective filing date of the claimed invention(s) absent any evidence to the contrary. Applicant is advised of the obligation under 37 CFR 1.56 to point out the inventor and effective filing dates of each claim that was not commonly owned as of the effective filing date of the later invention in order for the examiner to consider the applicability of 35 U.S.C. 102(b)(2)(C) for any potential 35 U.S.C. 102(a)(2) prior art against the later invention.

Claim 42-80 is/are rejected under 35 U.S.C. 103 as being unpatentable over Lutz (US Patent Pub. 2012/0282207) in view of Wang et al. (Green Chem., 2012, 14, pp 758-765, disclosed by third-party) and Wright et al. (WO 2016/007196).

Lutz discloses an antimicrobial compositions and a product comprising a preservative effective amount of the said composition (Abstract). Lutz discloses household; fabric care and pharmaceutical products ([0043] and claim 25). Lutz discloses the composition comprises caprylyl glycol, which meets the limitation of 1,2-octanediol, pentylene glycol, hexylene glycol, and any combination thereof (reference claim 7). Lutz discloses the composition further comprises an antimicrobial mixture (reference claim 1).

Lutz differs from the instant claims insofar as it does not disclose the 1,2-alkanediol as “bio”.

Wang et al. disclose conversion of microcrystalline cellulose to 1,2-alkanediols. Wang et al. disclose due to the diminishing fossil fuel reservoirs together with the ever-deteriorating environment associated with the worldwide fossil-fuel-based industry, it is imperative to make use of alternative feedstock for the production of chemicals and fuels (p. 758, Introduction).

It would have been obvious to one of ordinary skill in the art to have used “bio” based 1,2-alkanediols in the formulation of Lutz motivated by the desire preserve the environment as taught by Wang et al.

Lutz differs from the instant claims insofar as it does not disclose a bio-1-alkene regioselectivity of at least about 80%.

Wright disclose formation of bio-alkanes with 90-99% selectivity ([54]).

It would have been obvious to have formed bio-alkanes with 90-99% selectivity since they are known as taught by Wright.

Conclusion

Any inquiry concerning this communication or earlier communications from the examiner should be directed to NANNETTE HOLLOMAN whose telephone number is (571)270-5231. The examiner can normally be reached on Monday-Friday 9am-6pm.

Examiner interviews are available via telephone, in-person, and video conferencing using a USPTO supplied web-based collaboration tool. To schedule an interview, applicant is encouraged to use the USPTO Automated Interview Request (AIR) at http://www.uspto.gov/interviewpractice.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Frederick Krass can be reached on (571)272-0580. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see https://ppair-my.uspto.gov/pair/PrivatePair. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a

USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

/NANNETTE HOLLOMAN/
Primary Examiner, Art Unit 1612

Notice of References Cited	Application/Control No. 16/262,902	Applicant(s)/Patent Under Reexamination Burgo et al.	
	Examiner NANNETTE HOLLOMAN	Art Unit 1612	Page 1 of 1

U.S. PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Name	CPC Classification	US Classification
*	A	US-20120282207-A1	11-2012	Lutz; Patrick Jay	C10L10/00	424/70.1
	B					
	C					
	D					
	E					
	F					
	G					
	H					
	I					
	J					
	K					
	L					
	M					

FOREIGN PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Country	Name	CPC Classification
	N	WO-2016007196-A1	01-2016	WO	KAMBAM PAVAN	C10G3/44
	O					
	P					
	Q					
	R					
	S					
	T					

NON-PATENT DOCUMENTS

*		Include as applicable: Author, Title Date, Publisher, Edition or Volume, Pertinent Pages)
	U	
	V	
	W	
	X	

*A copy of this reference is not being furnished with this Office action. (See MPEP § 707.05(a).)
Dates in MM-YYYY format are publication dates. Classifications may be US or foreign.

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

CPC - Searched*		
Symbol	Date	Examiner

CPC Combination Sets - Searched*		
Symbol	Date	Examiner

US Classification - Searched*			
Class	Subclass	Date	Examiner

* See search history printout included with this form or the SEARCH NOTES box below to determine the scope of the search.

Search Notes		
Search Notes	Date	Examiner
PALM Inventor Search	09/13/2020	NH
EAST Search	09/13/2020	NH
NPL Search	09/13/2020	NH

Interference Search			
US Class/CPC Symbol	US Subclass/CPC Group	Date	Examiner

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

EAST Search History

EAST Search History (Prior Art)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
L2	7	(("8912373") or ("9856461") or ("4234752") or ("20050065389") or ("20030065233") or ("20060161025") or ("20140066666")).PN.	US-PGPUB; USPAT; USOCR	OR	OFF	2020/09/13 13:37
S1	937	((("BURGO") near3 ("Rocco")) OR (("WRIGHT") near3 ("Michael")) OR (("MOSSER") near3 ("Gary")) OR (("FEVOLA") near3 ("Michael"))).INV.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	OFF	2020/07/07 19:12
S2	378	(bio with alkene) bioalkene	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/07/07 19:19
S3	6,577	regioselectivity	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/09 14:30
S4	13	regioselectivity same alkanediol	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/09 14:30
S5	7	regioselectivity same (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/09 14:47
S6	1	("7385092").PN.	USPAT; USOCR	OR	OFF	2020/09/09 14:53
S7	2	wo-2016007196-$.did.	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	OFF	2020/09/11 11:19
S8	6	alkanediol same (household and pharmaceutical and cosmetic)	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:25
S9	3	(household and pharmaceutical and cosmetic) same (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26
S10	336	(household and pharmaceutical and cosmetic) and (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26

S11	6	((household and pharmaceutical and cosmetic) and (octanediol or (caprylyl adj glycol))).clm.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26
S12	11	((household and pharmaceutical and cosmetic) and (octanediol or ((caprylyl or hexylene or decylene) adj glycol) or alkanediol)).clm.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:47
S13	946	((("BURGO") near3 ("Rocco")) OR (("WRIGHT") near3 ("Michael")) OR (("MOSSER") near3 ("Gary")) OR (("FEVOLA") near3 ("Michael"))).INV.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	OFF	2020/09/11 19:14
S14	3	S13 and alkanediol	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/09/11 19:18

EAST Search History (Interference)

<This search history is empty>

9/13/2020 2:14:43 PM
C:\Users\nholloman\Documents\EAST\Workspaces\16262902.wsp

Doc Code:IDS.3P
Document Description: Third-Party Submission Under 37 CFR 1.290

PTO/SB/429(08-12)
Approved for use through 07/31/2015. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

U.S. PATENTS

Cite No	Patent Number	Kind Code[1]	Issue Date (YYYY-MM-DD)	First Named Inventor

U.S. PATENT APPLICATION PUBLICATIONS

Cite No	Publication Number	Kind Code[1]	Publication Date (YYYY-MM-DD)	First Named Inventor
1	20140066666	A1	2014-03-06	Oskar Koch

FOREIGN PATENTS AND PUBLISHED FOREIGN PATENT APPLICATIONS

Cite No	Foreign Document Number[3]	Country Code[2]	Kind Code[1]	Publication Date (YYYY-MM-DD)	Applicant, Patentee or First Named Inventor	T[5]
						☐

NON-PATENT PUBLICATIONS (e.g., journal article, Office action)

Cite No	Author (if any), title of the publication, page(s) being submitted, publication date, publisher (where available), place of publication (where available).	T[5]	E[6]

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

1	Wang et al., Efficient conversion of microcrystalline cellulose to 1,2-alkanediols over supported Ni catalysts, GREEN CHEM., 14:758-765 (2012)	☐	☐
2	Jong & Birmingham, Mushrooms as a Source of Natural Flavor and AromaCcompounds", Chapter 37 (pp. 345-366) in MUSHROOM BIOLOGY AND MUSHROOM PRODUCTS (Chinese University Press 1993)	☐	☐

STATEMENTS

The party making the submission is not an individual who has a duty to disclose information with respect to the above-identified application under 37 CFR 1.56.

This submission complies with the requirements of 35 U.S.C. 122(e) and 37 CFR 1.290.

☐ The fee set forth in 37 CFR 1.290(f) has been submitted herewith.

☒ The fee set forth in 37 CFR 1.290(f) is not required because this submission lists three or fewer total items and, to the knowledge of the person signing the statement after making reasonable inquiry, this submission is the first and the only submission under 35 U.S.C 122(e) filed in the above-identified application by the party making the submission or by a party in privity with the party.

☐ This resubmission is being made responsive to a notification of non-compliance issued for an earlier filed third-party submission. The corrections in this resubmission are limited to addressing the non-compliance. As such, the party making this resubmission: (1) requests that the Office apply the previously-paid fee set forth in 37 CFR 1.290(f), or (2) states that no fee is required to accompany this resubmission as the undersigned is again making the fee exemption statement set forth in 37 CFR 1.290(g).

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

Signature	/James Balls/		
Name/Print	James Balls	Registration Number (if applicable)	57703

Examiner Signature	/NANNETTE HOLLOMAN/	Date Considered	09/13/2020

*EXAMINER: Signature indicates all documents listed above have been considered,except for citations through which a line is drawn. Draw line through citation if not considered. Include a copy of this form with next communication to applicant. 1. If known, enter kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16. See MPEP 901.04(a). 2. Enter the country or patent office that issued the document, by two-letter code under WIPO standard ST.3. See MPEP 1851. 3. For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. 4. If known, enter the kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 . See MPEP 901.04(a). 5. Check mark indicates translation attached. 6. Check mark indicates evidence of publication attached.

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

Bibliographic Data

Application No: 16/262,902

Foreign Priority claimed: ○ Yes ◉ No

35 USC 119 (a-d) conditions met: ☐ Yes ☑ No ☐ Met After Allowance

Verified and Acknowledged: /NANNETTE HOLLOMAN/ Examiner's Signature | Initials

Title: Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same

FILING or 371(c) DATE	CLASS	GROUP ART UNIT	ATTORNEY DOCKET NO.
01/30/2019	424	1612	I0113-198U2
RULE			

APPLICANTS

Inolex Investment Corporation, Wilmington, DE,

INVENTORS

Rocco V. Burgo, Mullica Hill, NJ, UNITED STATES

Michael E. Wright, Hurricane, UT, UNITED STATES

Gary B. Mosser, Tabernacle, NJ, UNITED STATES

Michael J. Fevola, Belle Mead, NJ, UNITED STATES

CONTINUING DATA

This application has PRO of 62623985 01/30/2018

FOREIGN APPLICATIONS

IF REQUIRED, FOREIGN LICENSE GRANTED**

02/15/2019

STATE OR COUNTRY

UNITED STATES

ADDRESS

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036
UNITED STATES

FILING FEE RECEIVED

$2,780

Doc Code:IDS.3P
Document Description: Third-Party Submission Under 37 CFR 1.290

PTO/SB/429(08-12)
Approved for use through 07/31/2015. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

U.S. PATENTS

Cite No	Patent Number	Kind Code[1]	Issue Date (YYYY-MM-DD)	First Named Inventor
1	4234752		1980-11-18	YULIN WU

U.S. PATENT APPLICATION PUBLICATIONS

Cite No	Publication Number	Kind Code[1]	Publication Date (YYYY-MM-DD)	First Named Inventor
1	20050065389	A1	2005-03-24	De Bruyn
2	20030065233	A1	2003-04-03	Fuji
3	20060161025	A1	2008-06-10	Swern

FOREIGN PATENTS AND PUBLISHED FOREIGN PATENT APPLICATIONS

Cite No	Foreign Document Number[3]	Country Code[2]	Kind Code[1]	Publication Date (YYYY-MM-DD)	Applicant, Patentee or First Named Inventor	T[5]
1	1876162	EP	A1	2008-01-09	Furuya	☐

NON-PATENT PUBLICATIONS (e.g., journal article, Office action)

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

Cite No	Author (if any), title of the publication, page(s) being submitted, publication date, publisher (where available), place of publication (where available).	T[5]	E[6]
1	Swern et al., Hydroxylation and Epoxidation of Some 1-Olefins with Per-acids, JACS, 68:1504-1507 (1946)	☐	☐

STATEMENTS

The party making the submission is not an individual who has a duty to disclose information with respect to the above-identified application under 37 CFR 1.56.

This submission complies with the requirements of 35 U.S.C. 122(e) and 37 CFR 1.290.

☒ The fee set forth in 37 CFR 1.290(f) has been submitted herewith.

☐ The fee set forth in 37 CFR 1.290(f) is not required because this submission lists three or fewer total items and, to the knowledge of the person signing the statement after making reasonable inquiry, this submission is the first and the only submission under 35 U.S.C 122(e) filed in the above-identified application by the party making the submission or by a party in privity with the party.

☐ This resubmission is being made responsive to a notification of non-compliance issued for an earlier filed third-party submission. The corrections in this resubmission are limited to addressing the non-compliance. As such, the party making this resubmission: (1) requests that the Office apply the previously-paid fee set forth in 37 CFR 1.290(f), or (2) states that no fee is required to accompany this resubmission as the undersigned is again making the fee exemption statement set forth in 37 CFR 1.290(g).

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

Signature	/daniel altman/		
Name/Print	Daniel Altman	Registration Number (if applicable)	34115

Examiner Signature	/NANNETTE HOLLOMAN/	Date Considered	09/11/2020

*EXAMINER: Signature indicates all documents listed above have been considered,except for citations through which a line is drawn. Draw line through citation if not considered. Include a copy of this form with next communication to applicant. 1. If known, enter kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16. See MPEP 901.04(a). 2. Enter the country or patent office that issued the document, by two-letter code under WIPO standard ST.3. See MPEP 1851. 3. For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. 4. If known, enter the kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 . See MPEP 901.04(a). 5. Check mark indicates translation attached. 6. Check mark indicates evidence of publication attached.

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
14 January 2016 (14.01.2016)

WIPO | PCT

(10) International Publication Number
WO 2016/007196 A1

(51) International Patent Classification:
B01J 29/06 (2006.01) *C07C 5/32* (2006.01)
C07C 2/12 (2006.01) *C07C 11/167* (2006.01)

(21) International Application Number:
PCT/US2015/011371

(22) International Filing Date:
14 January 2015 (14.01.2015)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
62/021,651 7 July 2014 (07.07.2014) US

(71) Applicant: **COBALT TECHNOLOGIES, INC.** [US/US]; 500 Clyde Avenue, Mountain View, California 94043 (US).

(72) Inventors: **WRIGHT, Michael E.**; 500 Clyde Avenue, Mountain View, California 94043 (US). **MEYER, Andrew D.**; 500 Clyde Avenue, Mountain View, California 94043 (US). **SHAW, Andrew**; 500 Clyde Avenue, Mountain View, California 94043 (US). **BURNS-GUYDISH, Stacy M.**; 500 Clyde Avenue, Mountain View, California 94043 (US). **FETSCH, Erin E.**; 500 Clyde Avenue, Mountain View, California 94043 (US). **MUCHA, Jeanette M.**; 500 Clyde Avenue, Mountain View, California 94043 (US). **KAMBAM, Pavan**; 500 Clyde Avenue, Mountain View, California 94043 (US).

(74) Agents: **JACOBSON, Jill A.** et al.; Wilson Sonsini Goodrich & Rosati, c/o Cobalt Technologies, Inc., 650 Page Mill Road, Palo Alto, California 94304 (US).

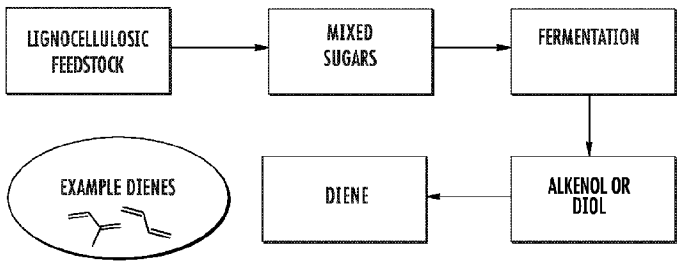
(81) Designated States *(unless otherwise indicated, for every kind of national protection available)*: AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IR, IS, JP, KE, KG, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States *(unless otherwise indicated, for every kind of regional protection available)*: ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

Published:

— *with international search report (Art. 21(3))*

(54) **Title**: BIOMASS CONVERSION TO BUTADIENE



NON-FOOD BIOMASS CONVERSION TO DIENES

FIG. 1

(57) **Abstract**: The present invention relates to the fermentation of lignocellulosic sugars to afford crotyl alcohol that is then separated and converted to butadiene using a dehydration catalyst and a method and apparatus for converting alkenols or diols to dienes with high selectivity and conversion. The dienes are useful in preparing synthetic rubber, copolymers, and fuels.

WO 2016/007196 A1

9/13/2020, EAST Version: 4.1.7.1

BIOMASS CONVERSION TO BUTADIENE

CROSS-REFERENCE TO RELATED APPLICATIONS

[01] This application claims the benefit of U.S. Provisional Application No. 62/021,651, filed on July 7, 2014, which is incorporated herein by reference in its entirety.

FIELD OF THE INVENTION

[02] The invention generally relates to conversion of biomass to dienes, especially butadiene, by dehydration of alkenols or diols. The dienes, including butadiene, are useful in the preparation of synthetic rubbers, plastics, fuels, and additional chemical intermediates incorporated into valuable commercial products.

BACKGROUND

[03] The energy and cost-efficient conversion of non-food biomass to strategic chemical intermediates is one major step in securing a sustainable future. Lignocellulosic materials and the mixed sugars (C5 and C6) that can be obtained from this non-food source (*e.g.*, corn stover, a waste product from food production) are an ideal feedstock for anaerobic fermentations of certain bacteria. Of particular value are *Clostridium* species that possess a robust and naturally-occurring ability to ferment C5 and/or C6 sugars into many useful products.

[04] The acetone, butanol, and ethanol (ABE) biosynthetic pathways in *Clostridium* include well-established bio-catalytic steps where the key intermediates in the pathway are well-characterized (Fig. 8). Interestingly, in the *Clostridium* ABE process/pathway to bio-1-butanol, a key intermediate is crotonyl-CoA (Fig. 9).

[05] A number of renewable butadiene processes have been proposed. These include construction of metabolic pathways to produce butadiene or intermediates thereof in a host organism. This requires introduction of multiple exogenous polynucleotides encoding multiple enzymes into a host organism. This in turn requires subsequent optimization of the expression of the multiple introduced genes and co-optimization of the kinetic profiles of the expressed genes to maximize flow of substrate to product. This is very time-consuming and has a low probability of success. Moreover, host organisms that produce butadiene will require the design and deployment of new fermenter types, which is expensive and introduces additional risk. Therefore, there is a need in the art to develop strategies for

renewable butadiene production that require minimal genetic manipulation of a host organism, minimal additional capital expenditure and minimal risk.

[06] α-Olefins are useful intermediates in preparing dienes, including 1,3-butadiene, diesel and jet/turbine fuels, and polymers. 1-Butene is an especially useful precursor to 1,3-butadiene, which is used in preparing synthetic rubber and other useful polymeric elastomers. α-Olefins are also useful in preparing poly-α-olefins (PAOs) and copolymers with ethylene to form low-density plastics and elastomeric materials. Renewable α-olefins are useful in preparing the corresponding renewable products, including renewable fuels, polymers, elastomers, PAOs, and other chemical intermediates. Currently, the production of ethylene from natural gas is rapidly replacing the steam cracking of naphthas and as a consequence, the four-carbon feedstocks (byproducts like C4-raffinate, a mixture of butane, 1-butene, and 2-butenes) are rapidly shrinking in availability and rising in cost. Thus, the traditional source of petroleum-based and non-renewable C4-feedstock typically used for preparing butadiene is vanishing. It is this shortage of the petroleum feedstock that establishes a need for a renewable and sustainable pathway to butadiene.

[07] Obtaining α-olefins and dienes (*e.g.*, 1,3-butadiene) from renewable and sustainable resources requires a significantly different approach than currently used in petroleum processes. Since alcohols can be produced in large scale by fermentation processes, they are an attractive feedstock for α-olefin and diene preparations, provided they can be dehydrated at high conversion rates and with high selectivity. This requires achieving high selectivity in forming the alkene bonds and avoiding formation of carbon-oxygen double bonds (*i.e.*, ketones or aldehydes) or the cleavage of carbon-carbon single bonds.

[08] The conversion of oxygen-containing derivatives to butadiene has received considerable attention over the past seven-plus decades with several reviews written on the topic. One example is the article written by Egloff & Hulla [UOP, *Chem. Rev.* **1945**, *36(1)*, pp 63-141]. Perhaps the most outstanding results to date in fact go back to the work reported by Reppe et al. in 1943 [US patent **2310809**]. This work indicated that 1, 3-butanediol and/or 1, 4-butanediol could be doubly dehydrated over a combination of phosphoric acid, conjugate bases, and additional organic components to give butadiene in yields of greater than 90%. However, the WHSV is low relative to that acceptable for a viable industrial process and the lifetime of the catalyst is placed in weeks rather than years. The latter is a typical requirement for a viable commercial catalyst. Thus, there exists a need for a catalyst with sufficient activity, selectivity, and lifetime on stream.

[09] Over the last decade, work from the Sato laboratory at Chiba University, Japan has explored the dehydration of diols to alkenols, diols to butadiene, and the dehydration of alkenols to butadiene in significant detail. This work has focused on using inorganic oxide catalysts. One result of particular relevance is the work Ichikawa et al. reported [*J. Mol. Catal. A: Chem.* **2006**, *256*, pp 106-112] on the dehydration of petroleum-derived 2-buten-1-ol (*i.e.*, crotyl alcohol). The best inorganic oxide catalyst was based on a SiO_2-Al_2O_3 composition where the dehydration of crotyl alcohol gave a 76% conversion with a 93% selectivity (*i.e.*, 71% yield) (Fig. 10).

[10] Commercial production of bio-1-butanol has a rich history of successful large-scale production since the discovery by Louis Pasteur in 1861 of bacteria that could ferment sugars to a mixture of acetone, 1-butanol, and ethanol (ABE). Since Pasteur's initial discovery of the ABE process, many advances have been made in the fermentation process to optimize bio-1-butanol production and to reduce ethanol and acetone co-production. Most notable are the successful efforts using *Clostridium* bacteria in commercial plants that can produce bio-1-butanol at a cost competitive to and even less than current petroleum-derived 1-butanol.

[11] Since fermentations are carried out in aqueous medium, separation of the fermentation products of interest from the multi-component mixture ("fermentation broth") of water, bacteria, media components, and other metabolic byproducts is energy- and time-intensive. In the case of bio-1-butanol, several methods have been reported for isolating the butanol component. One method that has found commercial success is use of a sparging gas (*e.g.*, carbon dioxide or steam) that carries the more volatile bio-1-butanol/water azeotrope away from the fermentation broth. Other more academic approaches involve pervaporation. In these cases, a selective membrane material is used that permits bio-1-butanol to pass through, thus leaving the bacteria and water behind. Regardless of the method, it is evident to those skilled in the art that removing the last traces of impurities and water are costly in energy and time. The available methods vary significantly in capabilities. However, water and impurities are a direct and unavoidable consequence of bio-alcohol fermentation processing.

[12] Ruwet et al. (*Bull. Soc. Chim.* **1987**, *96*, pp 281-292) discuss the problems associated with using a wet ABE bio-1-butanol feed in a dehydration reaction to afford a mixture of olefins. More recently, D'Amore et al. (US2008/0015395 A1) reported extreme difficulty in dehydrating aqueous solutions of 1-butanol using a variety of acid catalysts to afford a mixture of olefins and other oxygenated products (*e.g.*, ethers),

including high amounts of unreacted 1-butanol. Improved methods are needed to dehydrate bio-alcohols, in particular methods for dehydration of wet bio-alkenols to bio-dienes.

[13] Other alternative catalysts include silica and alumina solid phase catalysts as well as standard mineral acid catalysts. D'Amore et al. (US2009/0030239) disclose the contact of water and alcohols with acid catalysts to afford a mixture of alkene products. Mixtures are typical of acid catalysis and previous work in the field discloses that complicated mixtures of alkene products are formed. The lack of regioselectivity for terminal olefin occurs when isomerization to the more thermodynamically stable internal olefin occurs. In some cases the internal olefins can be formed directly via an acid catalyzed mechanism, typically denoted as *E1*. These results clearly show that dehydration results can be severally complicated by the addition of water in acid-catalyzed dehydrations of bioalcohols.

[14] Internal alkenes are far less effective in Ziegler-Natta polymerization chemistry and are not of direct use in preparing high-flashpoint diesel and jet fuels and for making lubricants. Interestingly, butadiene can be an effective route to bio-1-butene by selective hydrogenation (Fig. 11).

[15] As a feedstock, it is highly desirable to have a selective process that affords rapid and high conversion to a terminal-olefin (*e.g.*, bio-1-butene) and is accompanied by no carbon skeletal rearrangement (*e.g.*, to isobutylene). Furthermore, bio-dienes such as bio-1,3-butadiene can be oligomerized/polymerized using a variety of sophisticated methods known in the art. Products prepared from butadiene are useful intermediates in forming reactive end-capped oligomers, lubricants, and fuels. 1,3-butadiene is also a useful intermediate for preparing several 6-carbon chemical intermediates useful in preparing polymers like nylon6,6 (Fig. 12).

BRIEF SUMMARY OF THE INVENTION

[16] Systems and methods are described for catalytic conversion of alkenols or diols to diene products. In some embodiments, the alkenol or diol reactant feed contains water. For example, the alkenol or diol may be produced in a microbial fermentation, and the feed for the catalytic conversion to diene may contain aqueous liquid from the fermentation medium.

[17] In some embodiments, a microorganism is engineered such that it produces crotyl alcohol from lignocellulosic biomass. In other embodiments, a non-engineered microorganism produces crotyl alcohol.

[18] In some embodiments, microbial production of crotyl alcohol is achieved through the deletion or inactivation of the bcd gene from a *Clostridium* organism (*e.g.*, which results

in stopping formation of butyryl-CoA). In some embodiments this is achieved through the ability of the native promiscuous alcohol dehydrogenase (adhE, adhe2) to convert crotonyl-CoA to crotonaldehyde. In some embodiments this is achieved through the ability of the native promiscuous alcohol dehydrogenases (adhE, adhE2, bdhAB) to convert the crotonaldehyde to crotyl alcohol. In some embodiments the production of crotyl alcohol from an engineered *Clostridium* is enhanced through treatment of the engineered *Clostridium* (*e.g.*, engineered to delete or inactivate the bcd gene) with one or more chemical mutagen followed by selection in the presence of conditions that bias the selection towards strains with increased crotyl alcohol production.

[19] A strategy is described herein whereby a microorganism which produces *n*-butanol is converted into a microorganism that produces crotyl alcohol, by deletion, inhibition, and/or inactivation of the activity of one, and possibly two, enzyme(s) (*e.g.*, bcd). With this microorganism, crotyl alcohol can be produced by fermentation using existing fermenter designs. Said crotyl alcohol can rapidly and inexpensively be converted to butadiene using a novel catalyst, also disclosed herein, thereby minimizing capital expenditure.

[20] Other strategies are described herein in which non-engineered microorganisms (e.g., microorganisms that produce *n*-butanol) may produce crotyl alcohol by adjustment of culture conditions to favor production of this product.

[21] In some embodiments, catalyst systems and processes are provided to convert alcohols (*e.g.*, bio-alcohols) to the corresponding dienes (*e.g.*, bio-dienes). In some embodiments, the alcohols (*e.g.*, bio-alcohols) described herein are alkenols (*e.g.*, bio-alkenols) or diols, which are converted to the corresponding dienes (*e.g.*, bio-dienes). In additional embodiments, alkenols (*e.g.*, bio-alkenols) are converted to the corresponding -α,ω-dienes (*e.g.*, bio-α,ω-dienes). In certain embodiments, 4-carbon alkenols (*e.g.*, bio-alkenols) are converted to 1,3-butadiene (*e.g.*, bio-1,3-butadiene). In specific embodiments, crotyl alcohol is converted to 1,3-butadiene. In some embodiments the feedstock (alcohol (*e.g.*, bio-alcohol), alkenol (*e.g.*, bio-alkenol), 4-carbon alkenol (*e.g.*, bio-alkenol), crotyl alcohol, or diol) contains from about 0.1 wt-% to about 90 wt-% water and, using systems and processes disclosed herein, delivers a product stream containing the corresponding diene (*e.g.*, bio-diene) and water. In some embodiments, the diene (*e.g.*, bio-diene) is produced with about 90% to about 99% chemical selectivity. In some embodiments, a single pass over the catalyst system affords a chemical conversion of greater than about 95%, about 98%, or about 99%. In some embodiments, the diene (*e.g.*, bio-diene) is

produced employing WHSV values greater than about 0.3 hr^{-1}, *e.g.*, greater than about 1 and less than about 10 h^{-1}, or about 0.3 hr^{-1} to about 10 hr^{-1}. In some embodiments of the methods disclosed herein, at least one solid phase catalyst is maintained at a temperature of about 200 °C to about 440 °C, or about 200 °C to about 320 °C, with the feed a solution containing alkenol (*e.g.*, bio-alkenol) or diol (*e.g.*, bio-diol) and water at a pressure of about 1 psig to about 1000 psig to produce a diene (*e.g.*, bio-diene) in high chemical selectivity at high alcohol conversion. In one embodiment, bio-crotyl alcohol that is obtained from a fermentation broth (*e.g.*, after removal of minor acidic contaminants) is dehydrated to bio-1, 3-butadiene in a yield of greater than about 80%, or greater than 85%, or greater than 90%.

[22] In one aspect, a method is provided for dehydrating an alkenol or a diol to a diene, including: contacting and heating the alkenol or diol with at least one catalyst that includes an inorganic support, one or more promoter(s) selected from one or more Group 1 element(s), one or more Group 2 element(s), and combinations thereof, and one or more buffer-adjusted anion(s) at a WHSV at least about 0.3 hr^{-1}, thereby producing a diene product that includes one or more diene(s) at a one-pass yield greater than about 72 mol%, or greater than about 73 mol%. In some embodiments, the WHSV is about 0.3 hr^{-1} to about 10 hr^{-1}. In some embodiments, the alkenol or diol is in a mixture with about 1 ppm to about 93 wt% water, or about 15 wt% to about 70 wt% water. In some embodiments, the selectivity of diene production is at least about 75%, at least about 80%, at least about 85%, at least about 90%, for example, greater than 95%, or about 90% to about 99%.

[23] In some embodiments, the alkenol is a bio-alkenol. In one embodiment, the alkenol is crotyl alcohol. In an embodiment, the crotyl alcohol is bio-crotyl alcohol. In some embodiments, the alkenol is a C_4 to C_6 alkenol. In some embodiments, 1,3-butadiene is produced in a method for dehydrating an alkenol or diol as disclosed herein.

[24] In some embodiments, the inorganic support includes γ-alumina, for example, in the form of an extrudate. In some embodiments, the catalyst is further modified with at least one organosilane, for example, an alkoxysilane, such as a chlorosilane.

[25] In some embodiments of the catalytic dehydration methods disclosed herein, at least one carrier gas is provided, for example, but not limited to, nitrogen and/or argon, or a mixture thereof.

[26] In some embodiments of the catalytic dehydration methods disclosed herein, heating is performed at a temperature of about 200° C to about 400° C. In some embodiments, the temperature is about 250° C to about 320° C.

[27] In some embodiments, the method further includes separating water from the diene product(s), thereby producing dry diene product(s).

[28] In some embodiments, the product of the dehydration reaction is 1,3-butadiene, and the method further includes contacting the 1,3-butadiene product with a second catalyst under conditions sufficient to produce 1-butene. In some embodiments, the 1-butene is contacted with a third catalyst, *e.g.*, a Ziegler-Natta catalyst, a mesoporous oligomerization catalyst, or an acid catalyst, under conditions sufficient to produce oligomers. In some embodiments, the oligomers are used to produce a fuel, *e.g.*, a diesel fuel or a jet fuel. In some embodiments, a diesel fuel is produced that has a flashpoint of about 38° C to about 100° C, a Cetane rating of about 45 to about 60, and aromatic content of less than about 0.5 wt-%. In some embodiment, a jet fuel is produced that has a flashpoint of about 38° C to about 100° C, a cold flow viscosity of less than about 8.0 cSt at -20 C, and aromatic content of less than about 0.5 wt-%.

[29] In some embodiments, the promoter(s) is(are) selected from Mg, Ca, Sr, Ba, and combinations thereof. In one embodiment, the promoter is Ca. In some embodiments, the buffer-adjusted anion is selected from SO_4^{-2}, $H_2PO_4^{-1}$, and mixtures thereof.

[30] In another aspect, a method is provided for dehydrating an alkenol or a diol to a diene, including: contacting and heating the alkenol or diol with at least one catalyst that includes a calcium-modified inorganic support and one or more buffer-adjusted anion(s), thereby producing a diene product that includes one or more diene(s) at a one-pass yield greater than about 80 mol%. In some embodiments, diene product(s) is(are) produced with greater than about 90 mol% selectivity and greater than about 90 mol% conversion. In some embodiments, the alkenol is a bio-alkenol. In one embodiment, the alkenol is crotyl alcohol, *e.g.*, bio-crotyl alcohol. In some embodiments the alkenol is a C_4 to C_6 alkenol. In some embodiments, the diene product is 1,3-butadiene. In some embodiments, the alkenol is in a mixture with about 1 ppm to about 93 wt-% water. In some embodiments, the contacting of the alkenol or diol with the at least one catalyst is performed at a WHSV of about 0.3 hr^{-1} to about 10 hr^{-1}. In some embodiments, the catalyst is modified by treatment with at least one organosilane.

[31] In another aspect, a method is provided for dehydrating crotyl alcohol to a diene product, including: contacting and heating the crotyl alcohol with at least one catalyst that includes an inorganic support, one or more promoter(s) selected from one or more Group 1 element(s), one or more Group 2 element(s), and combinations thereof, and one or more buffer-adjusted anion(s) at a WHSV at least about 0.3 hr^{-1}, thereby producing a diene

product that includes one or more diene(s) at a one-pass yield greater than about 73 mol%. In some embodiments, the Group 2 element is Ca. In some embodiments, the buffer-adjusted anion is selected from SO_4^{-2}, $H_2PO_4^{-1}$, and mixtures thereof. In some embodiments, the crotyl alcohol is in a mixture that includes about 1 ppm to about 93 wt-% water. In some embodiments, the mixture includes about 5 wt-% to about 70 wt-% water. In some embodiments, the crotyl alcohol is bio-crotyl alcohol. In some embodiments, the diene product is 1,3-butadiene. In some embodiments, the molar selectivity of diene production is greater than about 95%, about 90% to about 99%. In some embodiments, the inorganic support includes γ-alumina. In some embodiments, the catalyst is further modified by treatment with at least one organosilane, for example an alkoxysilane. In some embodiments, the method includes providing at least one carrier gas.

[32] In another aspect, catalyst compositions are provided herein for producing dienes from alkenols or diols with high selectivity, including: an inorganic support that is modified with at least one Group 1 element, at least one Group 2 element, or a combination thereof; one or more buffer-adjusted anion(s); and optionally, at least one organosilane. In some embodiments, the catalyst composition includes at least one Group 2 element selected from Mg, Ca, Sr, and Ba. In one embodiment, the Group 2 element is Ca. In some embodiments, the buffer-adjusted anion is selected from SO_4^{2-}, $H_2PO_4^{-1}$, and mixtures thereof. In some embodiment, the buffer-adjusted anion is $H_2PO_4^{-1}$, wherein $H_2PO_4^{-1}$ is a component of a mixture that further optionally includes one or more additional anions selected form HPO_4^{-2}, and PO_4^{-3}, and one or more additional charge balancing counter cations selected from H^+, Li^{+1}, Na^{+1}, K^{+1}, Mg^{+2}, Ca^{+2}, Sr^{+2}, and Ba^{+2}, and combinations thereof. In some embodiments, the catalyst composition includes an organosilane, for example, diethoxydiphenylsilane. In some embodiments, the inorganic support includes γ-alumina, silica, titanium oxide, zinc oxide, zinc aluminate, or zirconium oxide.

[33] In some embodiments, catalyst compositions disclosed herein contain an inorganic support modified by a Ca^{+2} compound. In one embodiment, modification may be accomplished via treatment of the support with a mixture of calcium compounds, for example calcium bis(hydrogensulfate) and calcium sulfate, dissolved in a solvent. In some embodiments Ca^{+2} is the modifier and the Ca^{+2} compound is calcium bis(hydrogensulfate) [$Ca(HSO_4)_2$]. The solvent may be water, or an organic solvent, or a mixture of the two. In some embodiments, a solvent is used that is capable of completely dissolving the Ca^{+2} compounds used for preparation of the catalyst. In some embodiments, an "Incipient

Wetness Impregnation" (IWP) process is used for catalyst preparation. In one embodiment, the catalyst composition includes Ca^{+2} (as wt-% of the modified inorganic support) at about 0.8 to about 6.0 wt-%. In one embodiment, γ-alumina is used as the inorganic support. In some embodiments, γ-alumina is modified with binder, physically formed (*e.g.*, extruded) to a preselected shape (*e.g.*, rods), calcined, and then modified with a mixture of Ca^{+2} compounds as noted above in this paragraph. In some embodiments, calcium is preferred over Group 1 metals such as sodium and potassium when used at similar wt-% loading values.

[34] In one aspect, a method of making a catalyst composition described herein is provided. The method includes: a) modifying an inorganic support with at least one Group 2 element and at least one buffer-adjusted anion (e.g., a buffer mixture of anions) to produce a first modified inorganic support, wherein the first modified inorganic support includes the at least one Group 2 element and at least one buffer-adjusted anion; b) calcining the first modified inorganic support to produce a calcined first modified inorganic support; c) optionally contacting the calcined first modified inorganic support with at least one organosilane to produce a second modified inorganic support, wherein the second modified inorganic support includes the at least one Group 2 element, the at least one buffer-adjusted anion, and the at least one organosilane or a fragment thereof; and d) calcining the second modified inorganic support to produce the catalyst composition.

[35] In another aspect, a catalyst composition as described herein is provided, that is produced by a method that includes: a) modifying an inorganic support with at least one Group 2 element and at least one buffer-adjusted anion (*e.g.*, a buffer mixture of anions) to produce a first modified inorganic support, wherein the first modified inorganic support includes the at least one Group 2 element and at least one buffer-adjusted anion; b) calcining the first modified inorganic support to produce a calcined first modified inorganic support; c) optionally contacting the calcined first modified inorganic support with at least one organosilane to produce a second modified inorganic support, wherein the second modified inorganic support includes the at least one Group 2 element, the at least one buffer-adjusted anion, and the at least one organosilane or a fragment thereof; and d) calcining the second modified inorganic support to produce the catalyst composition.

[36] Promoter (*e.g.* Group 2 or Group 1 metal) modified and calcined catalysts may optionally be further modified by treatment with at least one organosilane to create new catalysts that are especially useful for the dehydration of alkenols (*e.g.*, bio-alkenols) or diols (*e.g.*, bio-diols) mixed with a high wt-% of steam. In some embodiments, buffer-

adjusted organosilane-modified catalysts afford high selectivity in forming diene (*e.g.*, bio-diene) products and can perform for extended periods of time on stream with little loss in selectivity.

[37] In some embodiments, methods disclosed herein for producing terminal olefins include using calcium or calcium-organosilane modified alumina catalysts, as described herein, for example, in isothermal continuous flow reactors or in a series of one or more adiabatic reactors. In some embodiments, some of the heat necessary for the dehydration reaction is carried into the reactor in the form of a gaseous diluent, such as steam. This creates a very water rich gas phase in the reactor making some embodiments of the methods disclosed herein beneficial for commercial applications.

[38] Some embodiments of the methods and systems described herein relate to fuels (*e.g.*, alternative and/or renewable fuels) and the production of dienes (*e.g.*, bio-dienes (*e.g.*, biodiesels)) that can be utilized to prepare a drop-in and full performance diesel fuel (*e.g.*, diesel biofuel), jet (*e.g.*, biojet) fuel, lubricant (*e.g.*, biolubricant), or other chemical products and/or intermediates useful in the production of commercial products.

[39] Embodiments also include such processes starting from cellulosic and/or hemicellulosic biomass material, as are shown schematically in FIG 1. Bio-alkenols or bio-diols can also be generated feeding a carbon-source of carbon dioxide and/or carbon monoxide to at least one biological organism. Typically the biological organism is suspended in water or it may be attached to at least one support system and then placed in contact with water and a carbon-source.

[40] In one aspect, methods are provided for dehydrating an alkenol (*e.g.*, bio-alkenol) to a diene (*e.g.*, bio-diene). In some embodiments, the method includes contacting and heating an alkenol (*e.g.*, bio-alkenol) with at least one catalyst that includes a calcium-modified (and optionally silanized) inorganic support at a WHSV of about 0.3 hr^{-1} to about 10 hr^{-1}, wherein the alkenol (*e.g.*, bio-alkenol) is in a mixture with about 1 ppm to about 93 wt% water, thereby producing the corresponding diene (*e.g.*, bio-diene) with greater than about 90% selectivity and greater than 98% conversion.

[41] In some embodiments of the methods disclosed herein, the alcohol (*e.g.*, bio-alcohol) is a C_4 to C_6 bio-alcohol. In one embodiment, the alcohol (*e.g.*, bio-alcohol) is butanediol (*e.g.*, 1,3-butanediol). The diene (*e.g.*, bio-diene) produced in some embodiments of the methods disclosed herein is 1, 3-butadiene. In some embodiments of the methods disclosed herein, the alcohol (*e.g.*, bio-alcohol) is a diol (*e.g.*, bio-diol) such as 2,3-butanediol or 1,3-butanediol.

[42] In some embodiments of the methods disclosed herein, the alcohol (*e.g.*, bio-alcohol) is an alkenol (*e.g.*, bio-alkenol) such as 2-butene-1-ol (*e.g.*, bio-2-buten-1-ol). For example, in some embodiments, the alkenol (*e.g.*, bio-alkenol) or diol (*e.g.*, bio-diol) may be a C_3 to C_{12} alcohol (*e.g.*, bioalcohol), *e.g.*, 1,12-decanediol (*e.g.*, bio-1,12-decanediol). The diene produced in some embodiments of the methods disclosed herein may be 1,3-butadiene (*e.g.*, bio-1,3-butadiene).

[43] In some embodiments of the methods disclosed herein, the 1-alkenol is an alcohol of general formula $R^1R^2CH{=}CHCH_2OH$, where R^1 and R^2 are independently H or a C_1 to C_{20} alkyl radical, or contain one or more aromatic rings containing exclusively carbon or a mixture of carbon and heteroatoms, or a nitrile, or an alkene, or a carboxylic acid, or ketone functional group, or a combination thereof.

[44] In some embodiments, the selectivity of diene (*e.g.*, bio-diene) production is greater than about 95%. In some embodiments, the selectivity of diene (*e.g.*, bio-diene) production is about 90% to about 99%. In some embodiments, the diene (*e.g.*, bio-diene) product includes $\leq$ about 5% unreacted 1-alcohol.

[45] In some embodiments, water is present in the feed reaction mixture at about 15 to about 70 wt%.

[46] In some embodiments, the inorganic support includes one or more phases of alumina (*e.g.*, γ-alumina), silica, titanium oxide, zirconium oxide, or zinc aluminate. In some embodiments, the inorganic support includes a combination of more than one of one or more phases of alumina, silica, titanium oxide, zirconium oxide, and zinc aluminate. The surface area of the inorganic support may be in the range of about 50 to about 400 m^2/g before modification and in the range of about 40 to about 440 m^2/g after modifications to form the catalyst.

[47] In some embodiments, the catalyst is further modified by treatment with at least one organosilane, for example, an alkoxysilane. In one embodiment, the organosilane is a chlorosilane. In one embodiment, the organosilane is diethoxydiphenylsilane. The organosilane may be dissolved in a solvent sufficient to form a homogeneous solution when combined with the organosilane and to wet the inorganic support.

[48] In some embodiments, the catalyst is further modified by treatment with at least one organosilane dissolved in an alcohol solvent. In some embodiments the alcohol solvent is *n*-propanol, 2-propanol, ethanol, or methanol. In one embodiment, the organosilane is diethoxydiphenylsilane and the alcohol solvent is ethanol. In another embodiment, the

organosilane is diethoxydiphenylsilane and the alcohol solvent is methanol. The alcohol solvent can contain from about 1 ppm water up to about 50 wt-% of the alcohol. For selected organosilane reagents with sufficient water solubility, water alone can be used to deliver the organosilane to the catalyst surface.

[49] In some embodiments of the methods of use of the catalysts, *e.g.*, for dehydration of alkenols or diols, at least one purge (*e.g.*, carrier) gas is provided, *e.g.*, nitrogen and/or argon.

[50] In some embodiments, the reaction mixture is heated to a temperature of about 200° C to about 440° C. In some embodiments, the temperature is about 260 °C to about 340 °C. In some embodiments, the temperature is about 250 °C to about 320 °C.

[51] In some embodiments, the method further includes separating water from 1, 3-butadiene product. In some embodiments, the method includes contacting the 1,3-butadiene product from which water has been removed with a second catalyst (hydrogenation type, *e.g.*, palladium on carbon) and hydrogen gas to form 1-butene (*e.g.*, bio-1-butene) in greater than 80% yield. Other more selective hydrogenation catalysts may also be used. The 1-butene (*e.g.*, bio-1-butene) is oligomerized with a third catalyst. In some embodiments, the third catalyst may be a Ziegler-Natta catalyst, a mesoporous oligomerization catalyst, or another catalyst that is capable of producing oligomers from dienes (*e.g.*, bio-dienes). In various embodiments, the oligomers may be used to produce a diesel fuel (*e.g.*, with a flashpoint of about 38 to about 100 °C, a Cetane rating of about 45 to about 60, and aromatic content of less than about 0.5 wt-%), a jet fuel (*e.g.*, with a flashpoint of about 38 to about 100 °C, a cold flow viscosity of less than about 8.0 cSt at -20 °C, and aromatic content of less than about 0.5 wt-%), or a lubricant (*e.g.*, with a viscosity of about 1 to about 10,000 cSt at 25 °C).

[52] In some embodiments, the method includes contacting and heating an alkenol (*e.g.*, bio-alkenol) or diol (*e.g.*, bio-diol) with at least one catalyst that includes a calcium-modified inorganic support and at least one buffer-mixture, thereby producing the corresponding diene (*e.g.*, bio-diene) with greater than about 90% selectivity and greater than about 90% conversion. Optionally, the alcohol (*e.g.*, bioalcohol) or diol (*e.g.*, bio-diol) is in a mixture with about 1 ppm to about 93 wt% water. Optionally, the catalyst is contacted with the alkenol at a WHSV of greater than 0.3 (*e.g.*, about 0.3 hr^{-1} to about 10 hr^{-1}). Optionally, the catalyst is further modified by treatment with at least one organosilane.

[53] In some embodiments, the method includes contacting and heating an alcohol (*e.g.*, bioalcohol, *e.g.*, alkenol (*e.g.*, bio-alkenol) or diol (*e.g.*, bio-diol)) with at least one catalyst

that includes an inorganic support that is modified by a Group 2 element, an acid component, and at least one organosilane, thereby producing the corresponding diene with greater than about 90% selectivity. For example, the Group 2 element may include at least one of Mg, Ca, Ba, and Sr. Optionally, the alkenol (*e.g.*, bio-alkenol) or diol (*e.g.*, bio-diol) is in a mixture with about 1 ppm to about 93 wt% water. Optionally, the catalyst is contacted with an alkenol (*e.g.*, bio-alkenol) or diol (*e.g.*, bio-diol) at a WHSV of greater than 0.3 (*e.g.*, about 0.3 hr^{-1} to about 10 hr^{-1}). In some embodiments Group 1 metals can be used in place of Group 2 elements. This includes Li, Na, and K and mixtures of Group 1 and Group 2 metals can be used in one embodiment.

[54] In some embodiments, dehydration of crotyl alcohol (a bio-alkenol) using the catalyst compositions and methods described herein provides 1,3-butadiene (*e.g.*, bio-1,3-butadiene) in greater than 90% yield, and, in some embodiments, 90-99% selectivity for 1,3-butadiene or 95-99% selectivity. In some embodiments, the catalyst can be used continuously for at least 1 to 3 months, or about 6 months and in some embodiments, up to about 12 months, or up to about 18 months, or up to about 24 months or up to about 30 months, or about 36 months, or up to 48 months while producing -1,3-butadiene without significant loss in selectivity for the terminal diene.

[55] In another aspect, the charge of the Group 2 element is balanced with at least one counter anion (*i.e.*, X^-, where "X" represents a generalized counter anion, not limited to a halide ion) that is a component of the mixture of anions, and charge balancing counter cations, used for the buffer-adjustment. In some embodiments, the pKa for at least one respective conjugate acid (*i.e.*, XH) is in the range of about 0 to about 3, or about 1.5 to about 2.5. The composition of the Group 2 element may use a mixture of counter anions having one or more pKa values for each respective conjugate acid. In one embodiment then the Group 2 element is matched with counter anions that effectively form a buffer-adjusted surface on the inorganic support. For example, a mixture containing both $Ca(HSO_4)_2$ and $Ca(SO_4)$ can be used to deliver a buffered-adjusted promoter to the inorganic oxide surface. Another embodiment employs a mixture of phosphates (H_3PO_4, and counter anions $H_2PO_4^{-1}$, and HPO_4^{-2}, and/or PO_4^{-3}), when modifying with calcium or other Group 2 metals. In another embodiment the mixture of phosphates can be used with one or more Group 1 metal cation promoters. In some embodiments, the Group 2 or Group 1 metal is added to the inorganic oxide support in the amount of about 0.3 to about 4 wt-%, and buffering counter anions are used to balance the positive charge of the Group 1 and/or Group 2 cations selected.

[56] In one aspect, a method is provided for producing crotyl alcohol, including growing a microorganism in a growth medium under fermentation conditions suitable for production of crotyl alcohol. In some embodiments, the microorganism expresses one or more native enzyme(s) that can produce crotyl alcohol from a substrate in the growth medium and/or via a metabolic pathway in the microorganism. In some embodiments, the enzyme is an alcohol and/or aldehyde dehydrogenase. In some embodiments, the enzyme is an acyl-CoA reductase. In some embodiments, the substrate is crotonaldehyde. In some embodiments, crotyl alcohol is produced via a series of biosynthetic intermediates that include crotonic acid, crotonyl-CoA and/or other intermediates in the butanol/butanoate pathway. In some embodiments, the native enzyme(s) is(are) enzyme(s) in a native butanoate/butanol, fatty acid, amino acid degradation, or related metabolic pathway in the microorganism. In some embodiments, the metabolic pathway includes one or more of an enzyme of a glycolytic or pentose phosphate pathway that converts glucose and/or xylose to acetyl-CoA, a thiolase enzyme that converts acetyl-CoA to acaetoacetyl-CoA, a 3-hydroxy butyryl CoA dehydrogenase enzyme that converts acetoacetyl-CoA to 3-hydroxybutyryl-CoA, a crotonase enzyme that converts 3-hydroxybutyryl-CoA to crotonyl-CoA, an aldehyde dehydrogenase enzyme that converts crotonyl-CoA to crotonaldehyde, and an alcohol dehydrogenase enzyme that converts crotonaldehyde to crotyl alcohol. In some embodiments, the microorganism is engineered to produce crotyl alcohol or for improved crotyl alcohol production in comparison to the parent microorganism from which it is derived.

[57] In some embodiments, the microorganism is a *Clostridium* strain. In some embodiments, the *Clostridium* microorganism expresses an enzyme, *e.g.*, a native enzyme, that converts crotonaldehyde to crotyl alcohol, for example, an alcohol and/or aldehyde dehydrogenase. In some embodiments, the *Clostridium* microorganism expresses an enzyme, *e.g.*, a native enzyme, that converts crotonyl-CoA to crotonaldehyde or crotyl alcohol, for example, an acyl-CoA reductase, for example, that converts crotonyl-CoA to crotyl alcohol without additional intervening enzymatic activities.

[58] In some embodiments, the growth medium includes one or more substance(s) that increase production of crotyl alcohol in comparison to a growth medium that does not comprise the one or more substance(s). In some embodiment, the one or more substance(s) includes one or more co-factors of enzyme(s) in a butanoate/butanol metabolic pathway in the microorganism, *e.g.*, Mn, Fe, NADPH, and/or NADP. In some embodiments, the one or more substance(s) modifies (*e.g.*, activates or inhibits) the activity of one or more

enzyme(s), *e.g.*, one or more native enzyme(s), in the microorganism, thereby increasing crotyl alcohol production. In one embodiment bcd is inhibited by the one or more substance(s) in the growth medium. In one embodiment, adhE and/or adh2 are activated by the one or more substance(s) in the growth medium. In one embodiment, bcd is inhibited by one or more substance(s) in the growth medium and adhE and/or adh2 are activated by one or more substance(s) in the growth medium. In some embodiments, the one or more substance(s) in the growth medium includes one or more divalent metal ion(s) (*e.g.*, from selected from Ca, Co, Zn, and Mn, and combinations thereof), iodoacetamide, myristoyl-CoA, N-ethylmaleimide, and/or palmitoyl-CoA. In some embodiments, the growth medium comprises carbohydrate molecules produced by hydrolysis of lignocellulosic biomass.

[59] In one aspect, an engineered microorganism is provided that produces a greater amount of crotyl alcohol than the parent strain from which it was derived. In some embodiments, the microorganism is a genetically engineered *Clostridium* strain. In some embodiments, the microorganism (*e.g.*, *Clostridium* strain) has been engineered to comprise a reduced level or activity of butyryl-CoA dehydrogenase in comparison to the parent strain from which it was derived. In some embodiments, the microorganism (*e.g.*, *Clostridium* strain) includes overexpression of one or more enzyme(s) of a crotyl alcohol biosynthetic pathway. In some embodiments, the one or more enzyme(s) of crotyl alcohol biosynthesis includes one or more of an enzyme of a glycolytic or pentose phosphate pathway that converts glucose and/or xylose to acetyl-CoA, a thiolase enzyme that converts acetyl-CoA to acaetoacetyl-CoA, a 3-hydroxy butyryl CoA dehydrogenase enzyme that converts acetoacetyl-CoA to 3-hydroxybutyryl-CoA, a crotonase enzyme that converts 3-hydroxybutyryl-CoA to crotonyl-CoA, an aldehyde dehydrogenase enzyme that converts crotonyl-CoA to crotonaldehyde, and an alcohol dehydrogenase enzyme that converts crotonaldehyde to crotyl alcohol. In some embodiments, the one or more enzyme(s) of crotyl alcohol biosynthesis includes at least one alcohol dehydrogenase. In some embodiments, the microorganism (*e.g.*, *Clostridium* strain) is an SiHybrid in which butyryl-CoA dehydrogenase activity is downregulated in comparison to the parent strain from which it was derived. In some embodiments, the microorganism (*e.g.*, *Clostridium* strain) includes a butyryl-CoA dehydrogenase that has been inactivated using a targetron gene knockout system.

[60] In some embodiments, methods are provided for producing crotyl alcohol in an engineered microorganism, *e.g.*, genetically engineered microorganism, *e.g.*, engineered *Clostridium* strain, as described herein, including growing the microorganism in a growth

medium under fermentation conditions suitable for production of crotyl alcohol. In some embodiments, the growth medium includes carbohydrate molecules produced by hydrolysis of lignocellulosic biomass.

BRIEF DESCRIPTION OF THE DRAWINGS

[61] FIG. 1 illustrates an overall process for converting biomass to dienes

[62] FIG. 2 illustrates a high process flow diagram showing the preparation of a promoter and buffer-adjusted dehydration catalyst.

[63] FIG. 3 illustrates a high process flow diagram showing the preparation of a promoter and buffer-adjusted dehydration catalyst that is also silanized.

[64] FIG. 4 illustrates a high level process flow diagram showing the dehydration of an alkenol or diol using the promoter and buffer-adjusted dehydration catalyst in a single reactor system to afford a diene.

[65] FIG. 5 illustrates a high level process flow diagram showing the dehydration of an alkenol or diol using the promoter and buffer-adjusted dehydration catalyst in a two reactor system (i.e. adiabatic) to afford a diene.

[66] FIG. 6 illustrates the acid-catalyzed isomerization of crotyl alcohol to 3-buten-2-ol, and isomerization of 3-buten-2-ol to crotyl alcohol, and dehydration of both crotyl alcohol and 3-buten-2-ol to 1,3-butadiene.

[67] FIG. 7 illustrates a high level process diagram for the conversion of a four carbon alkenol or diol to six carbon monomers.

[68] FIG. 8 illustrates schematically catalytic dehydration of an alkenol to a diene.

[69] FIG. 9 illustrates the chemical structure of crotonyl-CoA.

[70] FIG. 10 illustrates schematically catalytic dehydration of crotyl alcohol to 1,3-butadiene.

[71] FIG. 11 illustrates schematically catalytic hydrogenation of 1,3-butadiene to 1-butene.

[72] FIG. 12 illustrates schematically conversion of 1,3-butadiene to other useful products.

[73] FIG. 13 illustrates schematically catalytic dehydration of an alkenol to a diene.

[74] FIG. 14 illustrates schematically catalytic dehydration of a diol to a diene.

[75] FIG. 15 shows the effect of crotyl alcohol addition on *n*-butanol production in *Clostridium* fermentations.

[76] FIG. 16 shows the effect of crotyl alcohol addition on *n*-butanol production in *Clostridium* fermentations.

[77] FIG. 17 shows the effect of crotyl alcohol addition on total solvent production in *Clostridium* fermentations.

[78] FIG. 18 shows the effect of crotonic acid addition on glucose consumption over time in *Clostridium* fermentations.

[79] FIG. 19 shows consumption of crotonic acid over time in *Clostridium* fermentations.

[80] FIG. 20 shows the effect of crotonic acid addition on *n*-butanol production over time in *Clostridium* fermentations.

[81] FIG. 21 illustrates schematically biosynthetic pathways in *Clostridium*.

[82] FIG. 22 illustrates schematically biosynthetic production of crotyl alcohol.

[83] FIG. 23 illustrates schematically a system for genetic engineering.

[84] FIG. 24 shows production of crotyl alcohol from crotonaldehyde in a butanol-producing *Clostridium* strain.

[85] FIG. 25 shows production of *n*-butanol with crotonaldehyde spiking at different concentrations in a butanol-producing *Clostridium* strain.

[86] FIG. 26 shows production of crotyl alcohol from crotonaldehyde in a butanol-producing *Clostridium* strain.

DETAILED DESCRIPTION

[87] It is to be understood that the foregoing general description and the following detailed description are exemplary and explanatory only and are not to be viewed as being restrictive of the invention, as claimed. Further advantages of this invention will be apparent after a review of the following detailed description of the disclosed embodiments, which are illustrated schematically in the accompanying drawings and in the appended claims.

[88] Methods and catalyst compositions are provided herein for conversion of alkenols (*e.g.*, bio-alkenols) or diols (*e.g.*, bio-diols) to dienes (*e.g.*, bio-dienes). In methods disclosed herein, an aqueous mixture (*e.g.*, a mixture that contains at least one alkenol or diol and water) is contacted with a solid phase catalyst, which produces a product stream containing a diene with high selectivity (*e.g.*, greater than about 90%) and an amount of water equal to the input water (*e.g.*, steam) plus the stoichiometric amount of water (*e.g.*, steam) produced from the dehydration reaction, shown schematically in Fig. 13.

[89] Typically, the catalytic reaction is performed at an elevated temperature (*e.g.*, from about 200 °C to about 440 °C). Catalysts that may be used in the methods described herein include but are not limited to Ca^{+2} modified inorganic supports, such as Ca^{+2}-modified γ-alumina, and can optionally include an organosilane component. Catalyst compositions for use in the methods herein typically include at least one acid component. In some embodiments, Ca^{+2} is present at about 0.8 to about 4.0 wt-% of the modified inorganic support.

[90] Catalyst systems and methods of use are provided that effect highly specific regioselective dehydration of alcohols (*e.g.*, bio-alcohols), *e.g.*, alkenols (*e.g.*, bio-alkenols) and diols (*e.g.*, bio-diols), in water-containing mixtures, with retention of selectivity over extended periods of use, commonly referred to as Time On Stream (TOS).

Definitions

[91] The terms "terminal alkene," "α-olefin," "terminal olefin," and "1-olefin" are used interchangeably herein to refer to an alkene with a double bond between the terminal carbon (carbon at the end of a hydrocarbon chain) and the adjacent carbon in the hydrocarbon chain.

[92] The terms "internal alkene," and "internal olefin," are used interchangeably herein to refer to an alkene which is not terminal, *i.e.*, the carbon-carbon double bond is not at the end of the hydrocarbon chain. A "2-alkene" is one example of an "internal alkene".

[93] A "diol," of which 1,3-butanediol is one example, refers to a molecule having two hydroxyl groups (alcohol functional groups) attached to different carbon atoms in the molecule. In one embodiment, a "bio-diol," *e.g.*, "bio-1,4-butanediol" is a diol that is biologically produced (*e.g.*, via a microbial fermentation process). In another embodiment, a "bio-diol," *e.g.*, "bio-1,3-butanediol" is a diol that is produced from a starting molecule that is biologically produced (*e.g.*, via a microbial fermentation process).

[94] A "diene," of which 1,3-butadiene is one example, refers to a molecule having two olefinic (carbon-carbon double) bonds. The olefinic bonds may be isolated (*e.g.*, as in 1,5-hexadiene), conjugated (*e.g.*, as in 1,3-butadiene), or adjacent (allenic, *e.g.*, as in 1,2-butadiene). In one embodiment, a "bio-diene," *e.g.*, "bio-1,3-butadiene" is a diene that is produced from a starting molecule (*e.g.*, an alkenol) that is biologically produced (*e.g.*, via a microbial fermentation process). In some embodiments, a bio-olefin, bio-diene, or biobutadiene is produced from a bio-alkenol that has been produced in a microbial

fermentation process from cellulosic biomass (*e.g.*, lignocellulosic biomass) as a starting material.

[95] A "1-alcohol" or "terminal alcohol" refers, interchangeably, to an alcohol with a hydroxyl group attached to a terminal, primary carbon in the hydrocarbon chain (*i.e.*, a $-CH_2OH$ group).

[96] "*n*-Butanol" and "*n*-propanol" refer, respectively, to 1-butanol and 1-propanol.

[97] "Bio-*n*-butanol" or "bio-1-butanol" refers to *n*-butanol that is produced biologically (*e.g.*, via microbial fermentation). In some embodiments, the bio-1-butanol is produced in a microbial fermentation process from cellulosic biomass (*e.g.*, lignocellulosic biomass) as a starting material.

[98] An "alkenol" refers to an olefinic alcohol, *i.e.*, a molecule containing at least one olefinic (carbon-carbon double) bond and at least one alcohol functional group. A "bio-alkenol" is produced biologically (*e.g.*, via microbial fermentation). In some embodiments, the bio-alkenol is produced in a microbial fermentation process from cellulosic biomass (*e.g.*, lignocellulosic biomass) as a starting material. For example 2-buten-1-ol, if derived from biomass, would be considered a bio-alkenol.

[99] 2-Buten-1-ol refers to crotyl alcohol. No assignment of double bond geometry or stereochemistry is assumed or implied. As used herein, "2-buten-1-ol" or "crotyl alcohol" refers to any/all of: *cis*-2-buten-1-ol (*Z*-2-buten-1-ol), *trans*-2-butene-1-ol (*E*-2-buten-1-ol), or/and the combination of *cis*- and *trans*-2-buten-1-ol in any proportion. In some embodiments "bio-2-buten-1-ol" or "bio-crotyl alcohol" refers to biologically produced bio-2-butene-1-ol (crotyl alcohol), *e.g.*, via microbial fermentation, including via microbial fermentation of biomass (*e.g.*, lignocellulosic biomass). In other embodiments "bio-2-buten-1-ol" or "bio-crotyl-alcohol" refers to crotyl alcohol that is produced by a chemical dehydration of a bio-diol that is first obtained from a microbial fermentation process from biomass (*e.g.*, lignocellulosic biomass) feedstock. For example, bio-1,3-butanediol can be dehydrated to form bio-2-buten-1-ol.

[100] For alpha-olefins, alkenes, alcohols, solvents, alkenols, bio-alkenols, and all chemical reagents used in descriptions within this document, IUPAC and/or standard organic chemical nomenclature accepted by the American Chemical Society (ACS) takes priority and is used to the best ability to match common nomenclature.

[101] "Biofuel," "biolubricant," or "bio-1,3-butadiene" refers to a fuel, a lubricant, or 1,3-butadiene, respectively, that is produced from at least one biologically-produced starting molecule or at least one starting molecule in which at least one carbon atom is derived from

a biological material, for example, cellulosic (*e.g.*, lignocellulosic) biomass. Also the direct biological conversion of carbon dioxide and/or carbon monoxide is considered also defined as a viable route to bio-alkenols or bio-diols. In one embodiment, a biofuel, biolubricant, or bio-1,3-butadiene may be produced from a bio-alkenol that is produced by a biological process as described herein.

[102] A "bio-" prefix is used to indicate a molecule or class of molecules, for example bio-alkenes or bio-alkenols, that are produced from at least one starting molecule in which at least one carbon atom is derived from a biological material, for example, cellulosic (*e.g.*, lignocellulosic) biomass.

[103] "Regioselectivity" refers to selective formation of one olefin product over another. For example, formation of 90 mol-% terminal (1-olefin) and 10 mol-% internal (*e.g.*, 2-olefin) would be described as 90% regioselectivity for the terminal olefin product.

[104] "Selectivity" refers to product selectivity if not otherwise defined.

[105] "Chemical Yield" and "Yield" refer to are calculated by multiplying the conversion (percent) by the selectivity (percent). For example, if reaction A that has a 90% conversion with a 90% selectivity for product X, then product X has a 81% yield in reaction A.

[106] "Incipient Wetness Impregnation" (IWP) is a technique that is used to modify a support material by treating the solid material with a predetermined amount of a solution containing at least one desired modifier (*e.g.*, calcium acetate), sufficient to wet the solid surface and fill the pore-volume, this followed by removal of some or all of the solvent (*e.g.*, by some method of evaporation) thus depositing the modifier uniformly on the support material.

[107] "Weight Hourly Space Velocity" (WHSV) is determined by dividing the weight of alcohol feed delivered per hour by the weight of catalyst used. For example, if 1.5 kg/hr of terminal alcohol are fed to a reactor containing 500 grams of catalyst, the WHSV is 3.0 hr^{-1}. The alcohol feed rate, in units of WHSV, as defined herein, is independent of the amount of water, or other components that may be in the feed, or co-fed to the catalyst bed.

[108] "Buffer-Adjusted Anions" refers to a mixture of anions in a pH buffer solution. For example a sulfuric acid buffer will contain HSO_4^{-1} and SO_4^{-2} anions as part of the buffer solution. The required cations are chosen in the process of making the buffer solution.

[109] "Ph" herein refers to a phenyl (C_6H_5-) group.

[110] "Me" herein refers to an methyl (CH_3-) group.

[111] "Et" herein refers to an ethyl (CH_3CH_2-) group.

[112] "Ac" herein refers to an acetate (CH_3COO-) group.

[113] "EFT X" in Figs. 24-26 refers to elapsed fermentation time of X hours.

Catalyst compositions and methods of preparation

[114] One embodiment for the dehydration catalysts disclosed herein is an inorganic support modified by contact with one or more promoter-containing compound(s), *e.g.*, a Ca^{2+}-containing compound. In one embodiment, a promoter (*e.g.*, a calcium-containing compound) is delivered as a solution to the inorganic support using incipient wetness impregnation. In another embodiment, a promoter (*e.g.*, a calcium-containing compound) is delivered by spraying a promoter-containing (*e.g.*, calcium compound-containing) solution onto an inorganic support while the latter is carried on a belt system or is mixed in a rotating drum. In a further embodiment, vapor phase deposition is used to deposit promoter(s) on an inorganic support. Solvents used to prepare the promoter solution may be aqueous, or one or more organic solvent(s), or a mixture thereof. Nonlimiting examples of calcium-containing compounds that may be used for production of the catalysts described herein include $Ca(SO_4)$, $Ca(HSO_4)_2$, $Ca_3(PO_4)_2$, $Ca(HPO_4)$, or $Ca(H_2PO_4)_2$, any one of which may be available and used in more hydrated, or more dehydrated, or more condensed forms, than suggested by the chemical formulas cited here. Other calcium and Group 2 compounds, or Group 1 compounds, or Group 3 compounds, or Group 4 compounds, or rare earth compounds may be employed for this purpose as promoters.

[115] In addition to one or more promoter(s), the catalyst compositions disclosed herein include one or more "Buffer-Adjusted Anion(s)" (BAA). The selected anion, or mixture of anions, incorporated into the catalyst with the chosen promoter or mixture of promoters, improves the selectivity of the dehydration. The counter cations that accompany one or more BAA may include promoter(s) that are advantageous for the dehydration reaction, for example Ca^{+2}, or they can include a mixture of solely Group 2 or solely Group 1 metal cations, or a mixture of both Group 1 and Group 2 ions. In some embodiments, the BAA includes phosphate-containing ions or sulfate-containing ions, which have a well-established history of use in preparing aqueous buffer solutions. Nonlimiting examples of BAA include sulfate (SO_4^{-2}) and dihydrogenphosphate ($H_2PO_4^{-1}$) anions.

[116] In another embodiment, BAA is added as counter anion to the promoter using at least one Group 1 or Group 2 metal counter cation, in which case provision is also made to provide a buffering quantity of the conjugate acid of the BAA. For example, where the BAA is dihydrogenphosphate ($H_2PO_4^{-1}$), the conjugate acid (H_3PO_4), is also supplied, as exemplified by "Catalyst 4," described in Example 10, *infra*. The promoter metal ion

concentration may be maintained at about 0.5 wt-% to about 4 wt-% of the inorganic (*e.g.*, oxide) support. Suitable BAA include those anions wherein the pKa of the conjugate acid of the BAA falls in the range of zero to about 3. One non-limiting example of a suitable BAA for this embodiment is sulfate ion, whose conjugate acid, hydrogen sulfate ion (HSO_4^{-1}), has a pKa of about 2, between zero and about 3. Having selected SO_4^{-2} according to the criterion that the pKa of the conjugate acid falls within the range of zero to about 3, the conjugate acid (HSO_4^{-1}), and the corresponding free acid (H_2SO_4), in combination with the selected anion(s), may be used to prepare the catalyst.

[117] The BAA may be delivered I the form of its conjugate acid. For example, in a catalyst where sulfate (SO_4^{-2}) is among the selected BAA(s), the catalyst may be formulated using $NaHSO_4$ (where HSO_4^{-1} is the conjugate acid of SO_4^{-2}), as in "Catalyst 2," described in Example 8, *infra*. Regardless, for a selected BAA, the catalyst is formulated using a source of the conjugate acid of the BAA, or another acid with a pK1 within the range of zero to about 3. For example, in the case that the selected BAA is SO_4^{-2}, a source of the conjugate acid, HSO_4^{-1}, may be supplied, or H_3PO_4, another acid with a pKa within the range zero to about 3, may be supplied. The conjugate acid of the BAA may be supplied directly, or by a suitable combination of anions, and corresponding charge balancing cations. For example, where the BAA is SO_4^{2-}, the conjugate acid may be delivered directly as a HSO_4^{-1} salt (including Group 1 or Group 2 elements, such as $NaHSO_4$ or $Ca(HSO_4)_2$), or by a combination of H_2SO_4 and a SO_4^{2-} salt (including Group 1 or Group 2 elements, such as Li_2SO_4 or $CaSO_4$).

[118] In some embodiments the free-acid, for example, H_3PO_4 or H2SO4, is added as part of the BAA solution. Further, it is understood that, depending upon several factors, including the relative ratios of selected BAAs, the relative amounts of selected promotors, the surface properties of the selected inorganic support, and the composition of the diol or alkenol feed to the dehydration catalyst, during catalyst operation a portion of the BAAs present in the original catalyst formulation may be present as the corresponding free acid, *i.e.*, the counter cation(s) will be protons (H^+ ions). There is no requirement that all the BAAs, or their corresponding conjugate acids, are charge-balanced exclusively by promotor cations, either in the catalyst as-synthesized, as-formulated, as-charged to the dehydration reactor, or in-use. For example, even though the BAA SO_4^{-2} ion may be delivered using the sodium salt of the conjugate acid, HSO_4^{-1} ($NaHSO_4$), as described above, in which case the counter cation is a promoter (Group 1), alternatively the HSO_4^{-1} may be delivered as H_2SO_4 or $(NH_4)HSO_4$. It is understood and expected that a portion of the BAAs may be effectively

present in the buffered catalyst as the corresponding free acid, *e.g.*, H_2SO_4, if HSO_4^{-1} or SO_4^{-2} are among the anions in the catalyst formulation.

[119] In one embodiment of a catalyst composition disclosed herein, the inorganic support is γ–alumina, the Group I element is Na, the buffer-adjusted anion is sulfate (SO_4^{-2}), and the catalyst is formulated using the sodium salt of the conjugate acid of the buffer-adjusted anion, $NaHSO_4$.

[120] In one embodiment of a catalyst composition disclosed herein, the inorganic support is γ -alumina, the Group 2 element is Ca, the buffer-adjusted anion is dihydrogenphosphate ($H_2PO_4^{-1}$), and the catalyst is formulated using a mixture of the Ca salt of the buffer-adjusted anion [$Ca(H_2PO_4^{-1})_2xH_2O$] and the conjugate acid of the buffer-adjusted anion (H_3PO_4).

[121] In one embodiment of a catalyst composition disclosed herein, the inorganic support is γ -alumina, the Group 2 element is Ca, the buffer-adjusted anion is dihydrogenphosphate ($H_2PO_4^{-1}$), the catalyst is formulated using a mixture of the Ca salt of the buffer-adjusted anion [$Ca(H_2PO_4^{-1})_2xH_2O$] and the conjugate acid of the buffer-adjusted anion (H_3PO_4), and organosilane is diethoxydiphenylsilane, delivered to the catalyst from a solution in ethanol.

[122] In some embodiments, a promoter (*e.g.*, Ca^{+2}) buffer-adjusted mixture is dissolved in one or more solvent(s) to create a solution, the promoter (*e.g.*, Ca^{+2}) solution is placed in contact with the carrier (*e.g.*, ϒ-alumina), and the solvent(s) is (are) then removed, *e.g.*, with heat and/or reduced pressure. Those experienced in the methods of surface modification know that one technique to accomplish this is referred to as incipient wetness impregnation (IWP). IWP is typically followed by a calcination process of some time, as shown schematically in FIG 2. The inorganic support (**200**) is added to vessel **201** and then addition of the IWP solution **202**, which includes promoter (*e.g.*, Ca^{2+}) and BAA, is done in such a way as to provide even distribution. This can be done, for example, by adding solution **202** by a spraying method. Mechanical mixing in apparatus **203** can also be used facilitate even distribution of the promoter solution. The wetted material in **203** is then heated to remove some or all of the IWP solvent. This material **204** can be further heated in a second apparatus (**205**) to provide calcination of the catalyst. The calcination can be carried out in vessel **203** or **205** at temperatures of about 100 °C to about 900 °C, or about 200 °C to 600 °C, for a time period of about 1 hour to about 8 hours, or about 2 hours to 6 hours. The removal of the solvent can be done in air or under an inert atmosphere. In one embodiment the wetting solution is removed under a reduced pressure. The catalyst

removed from **203** or **205** can be sized by sifting or other methods common to solid phase catalyst processing, thus producing finished catalyst **206**. The concentration of Ca^{+2} in the solution is predetermined in such a way that a targeted amount of Ca^{+2} is deposited on the carrier (*e.g.*, alumina) surface. In some embodiments, the Ca^{+2} is deposited in the range of about 0.8 wt-% to about 4.0 wt-% or about 1.0 wt-% to 1.7 wt-% of the total catalyst {weight of Ca^{+2}/(weight of inorganic support + weight of Ca^{+2}) x 100 = wt-%}. Delivery of the calcium and the buffer-adjusted anions can also be made using other techniques such as, non-IW impregnation or, for example, vapor deposition followed by calcination in air or under an inert atmosphere. In some embodiments, all of the modifications are carried out on the final shape or sized support material.

[123] In some embodiments, the promoter-doped (*e.g.*, Ca^{+2}) and buffer-adjusted inorganic support (*e.g.*, γ-alumina) can be further modified by treatment with one or more organosilane(s). Treatment with organosilane(s) may provide a catalyst with improved stability for the dehydration of alcohols. In some embodiments, the organosilane(s) can be delivered to the inorganic support (*e.g.*, a Ca^{+2} and buffer-adjusted modified γ-alumina carrier) using IWP, as shown in a high level and general view in FIG 3. In this embodiment the calcium-modified buffer-adjusted inorganic support **206** is treated with the IWP solution **301** in such a way as to provide even distribution. This can be accomplished by spraying the organosilane(s) or a solution containing organosilane(s) **301** while mechanically mixing the promoted carrier in vessel **302**. The mixing process can greatly facilitate achieving an even distribution of the organosilane(s). However, it is not always necessary. The wetted material in apparatus **302** is then heated to remove some or all of the IWP solution and/or can be transferred to vessel **303** for further drying under ambient or reduced pressure. The catalyst (**304**) can be formulated, shaped, and sized by sifting or other methods common to solid phase catalyst processing. In one embodiment, a shaped or sized support (*e.g.*, **206**) may be used in the modification process, hence providing the catalyst **304** in an advantageous physical state (*e.g.*, shape or particle distribution).

[124] An organosilane solution **301** used to modify the carrier, *e.g.*, by IWP, can be made using water, one or more organic solvent(s), or a mixture of water and organic solvent(s) with or without additional additives. In one embodiment, the solvent is selected so that sufficient organosilane is soluble in the chosen solvent to deliver the required amount of organosilane to the solid by incipient wetness. Typical concentration of organosilane(s) when used in a IWP solvent is about 0.05 wt-% to about 4 wt-%, about 0.1 wt-% to about 0.2 wt-%, about 0.15 wt-% to 1.5 wt-%, or 0.3 wt-% to 1.3 wt-% total organosilane

concentration. In another embodiment, the organosilane(s) can be delivered in neat form, for example, as a liquid or in the vapor phase. Contact time for the neat organosilane(s) or organosilane-IWP solution with modified carrier may be about 5 minutes to about 6 hours, about 1 hour to about 6 hours, or about 2 hours to about 4 hours, thus allowing for complete dispersion and reaction, at or about ambient temperature (*e.g.*, about 20 °C to about 30 °C). Solvent(s) removal may be accomplished by the application of heat and/or reduced pressure. The typical final temperature reached is about 40 ° C to about 200 °C, about 300 °C, or about 400 °C. Removal of the solvent(s), if used, can be done in air or under an inert atmosphere (*e.g.*, nitrogen or argon).

[125] Typical organosilanes for use in the catalyst preparation methods disclosed herein include, but are not limited to, chlorosilanes, for example, Ph_2SiCl_2 and Ph_3SiCl. In some embodiments, alkoxysilanes may be used. For example, organosilanes such as Ph_3SiOEt and $Ph_2Si(OEt)_2$ can also be used to create a silanized-dehydration catalyst. Furthermore, R_3SiX, R_2SiX_2, and $RSiX_3$ organosilanes can be used, where R is a mixture of aryl and alkyl radicals and X represents a leaving group. The R-groups can be the same or a combination of alkyl, aryl, or organic functionalized substituents. Typical leaving groups include, but are not limited to, halide, alkyl carboxylate (*e.g.*, acetate), alkoxy (*e.g.* methoxy), or alkyl di- (*e.g.*, oxalate) and tri-carboxylates (*e.g.*, citrate).

[126] In some embodiments, water soluble organosilane(s) are employed in the silanization process (*e.g.*, treatment of the support with an organosilane solution and calcining). For these embodiments, the organosilane(s) may be soluble in water, for example, at levels of up to about 3 wt-%. The water solubility can be achieved by the use of hydrogen bonding groups attached to the silicon atom [*e.g.* $PhSi(OH)_3$] or by incorporation of a hydrogen bonding group or a dipolar functional group to one or more of the groups attached to the silicon atom. Examples could include, but are not limited to, an alkyl amine, ether, epoxide, or other dipolar or hydrogen bonding functional groups. In some embodiments, solubility of the organosilane(s) can be increased by heating a water solution to at least about 40 °C, about 50 °C, about 60 °C, or about 90 °C.

[127] The effect and benefit of silanization is dependent upon the carrier used, alkenol or diol feed, and the wt-% of water content added or contained in the alcohol feed. Typically, reaction conditions employing high water concentrations (*e.g.*, >50 wt-%) in the feed may exhibit the greatest benefit from silanization of the catalyst and afford very stable dehydration catalysts. Silanization of calcium promoted, or Group 1, or Group 2, or other promoted modified aluminas provides a catalyst that provides enhanced stability, which is

reflected in retention of selective diene (*e.g.*, bio-diene) production as disclosed herein. The process of silanization can be applied to aluminas (*e.g.*, γ-alumina) or other inorganic supports (*e.g.*, zinc aluminate) modified with Group 3, or Group 4, or Group 5, or Group 6, or Group 7, or Group 8, or Group 9, or Group 10 metals.

[128] Catalyst products **206** and **304** may be left as powders, for example, in the range of about 10 to about 1000 microns, or about 50 to about 200 microns, or they can be pelletized or shaped using techniques common to those skilled in the art. In some embodiments, pellets and shaped particles may provide advantages in controlling the pressure drop across the catalyst reactor bed. This may be important where physical process requirements require long reaction tubes or where low head-pressures are desirable. Pellets may be any shape or size. In some embodiments, pellet diameters are about 1/16 inch to about ¼ inch, with lengths about 1 to about 10 times the diameter, or about 2 to about 4 times the diameter. One embodiment includes first shaping, or pelletizing, or crushing with size selection (*e.g.* sieving), of the support, followed by the addition of at least one promoter as described herein.

Dehydration Methods

[129] The catalyst compositions described herein may be used to dehydrate alkenols (*e.g.*, bio-alkenols) or diols (*e.g.*, bio-diols), to produce diene (*e.g.*, bio-diene) products. In one embodiment, catalyst compositions disclosed herein may be used to dehydrate crotyl alcohol (*e.g.*, bio-crotyl alcohol) to 1,3-butadiene (*e.g.*, 1,3-bio-butadiene). In some embodiments, the catalyst compositions disclosed herein may be used to dehydrate alkenols or diols in the presence of water.

[130] In methods for dehydration of a alkenol or diol using the catalyst compositions disclosed herein, the water content in the initial alkenol (*e.g.*, bio-alkenol) or diol (*e.g.*, bio-diol) mixture (alkenol-water or diol-water mixture) that is fed to the catalyst can be from about 0.1 wt-% to about 93 wt-%, about 1 wt-% to about 70 wt-%, about 10 wt-% to about 50 wt-%, about 15 wt-% to about 19 wt-%, or about 17 wt-%. The water in the alkenol or diol feed can be contained in an alkenol-water mixture or diol-water mixture, or added using a second and separate feed, or may be a combination of the two {*e.g.*, total water in feed = (alkenol or diol + water mixture) + water feed}. In some embodiments, additional water is added to the alkenol-water or diol-water mixture prior to contact with the catalyst, thereby producing an alkenol or diol feed that contains about 30 wt-% to about 93 wt-% of water, or about 45 wt-% to about 70 wt-% of water. The use of high water content in the alkenol or

diol feed is especially well tolerated with catalysts prepared as disclosed herein. In some embodiments, the alkenol or diol feed in contact with the catalyst (*e.g.*, optionally including additional water added to the alkenol-water or diol-water mixture prior to entering the reactor and/or water added separately into the reactor) includes about 1 wt-% to about 70 wt-% water for optimum production of diene (*e.g.*, bio-diene), *e.g.*, 1,3-butadiene (*e.g.*, bio-1, 3-butadiene). In some embodiments, the alkenol or diol feed includes >90 wt-% water. In some embodiments, the water content in the alkenol or diol feed is about 1, about 5, about 10, about 15, about 20, about 25, about 30, about 35, about 40, about 45, about 50, about 55, about 60, about 65, about 70, about 75, about 80, about 85, or about 93 wt-% or more. In some embodiments, the water content in the alkenol or diol feed is about 15 to about 20 wt-%, about 15 to about 70 wt-%, or about 50 to about 80 wt-%. The diene (*e.g.*, bio-diene) product stream of the reaction may contain an amount of water as described above in this paragraph (*e.g.*, about 0.1 wt-% to about 93 wt-%), or values or subranges described above that are greater than the stoichiometric amount of water that is produced in the dehydration reaction.

[131] Catalysts disclosed herein may dehydrate wet alkenol (*e.g.*, bio-alkenol) or diol (*e.g.*, bio-diol) mixtures that contain large amounts of water. In some embodiments, steam may be added prior to contact with the catalyst to afford high selectivity for the diene (*e.g.*, bio-diene) dehydration product. Furthermore, in some embodiments, a high selectivity may be maintained for extended periods of time on stream, for example, about 1 month to about 3 months, about 6 months, about 12 months, about 18 months, about 24 months, about 30 months, about 36 months, about 42 months, about 48 months, or longer.

[132] Catalysts disclosed herein may also dehydrate an alkenol (*e.g.*, bio-alkenol) or diol (*e.g.*, bio-diol) mixture that contains water at levels as low as 1 ppm to 1000 ppm where equal performance of catalyst to afford diene (*e.g.*, bio-diene) product is observed throughout the entire range of low water content and the catalyst can maintain high selectivity for extended periods of time on stream, for example, about 1 month to about 3 months, about 6 months, about 12 months, about 18 months, about 24 months, about 30 months, about 36 months, about 42 months, about 48 months, or longer.

[133] In one embodiment of a method for dehydrating an alkenol or diol to a diene as described herein, the alkenol is crotyl alcohol, the diene is 1,3-butadiene, the inorganic support is γ-alumina, the Group 1 element is Na, the buffer-adjusted anion is sulfate (SO_4^{2-}),

and the catalyst is formulated using the sodium salt of the conjugate acid of the buffer-adjusted anion, $NaHSO_4$.

[134] In one embodiment of a method for dehydrating an alkenol or diol to a diene as described herein, the alkenol is crotyl alcohol, the diene is 1,3-butadiene, the inorganic support is γ -alumina, the Group 2 element is Ca, the buffer-adjusted anion is dihydrogenphosphate ($H_2PO_4^{-1}$), and the catalyst is formulated using a mixture of the Ca salt of the buffer-adjusted anion [$Ca(H_2PO_4^{-1})_2xH_2O$] and the conjugate acid of the buffer-adjusted anion (H_3PO_4).

[135] In one embodiment of a method for dehydrating an alkenol or diol to a diene as described herein, the alkenol is crotyl alcohol, the diene is 1,3-butadiene, the inorganic support is γ -alumina, the Group 2 element is Ca, the buffer-adjusted anion is dihydrogenphosphate ($H_2PO_4^{-1}$), the catalyst is formulated using a mixture of the Ca salt of the buffer-adjusted anion [$Ca(H_2PO_4^{-1})_2xH_2O$] and the conjugate acid of the buffer-adjusted anion (H_3PO_4), and organosilatne is diethoxydiphenylsilane, delivered to the catalyst from a solution in ethanol.

[136] An example of a catalytic alkenol or diol dehydration process as disclosed herein is shown schematically in FIG 4. An alkenol (*e.g.*, bio-alkenol) or diol (*e.g.*, bio-diol) feed **400** is introduced to the vaporization/mixing vessel (**401**) to which a water feed is added at some preselected amount. The output of **401** is transferred to vessel **403,** which contains the dehydration catalyst (*e.g.*, **206** or **304** prepared above) at an elevated temperature, for example, about 200 °C to about 440 °C, or about 250 °C to about 420 °C. Optionally, a purge (*e.g.*, carrier) gas, *e.g.*, an inert gas such as argon and/or nitrogen, may used to control dwell time in the reactor chamber. Temperature may be modified to optimize diene (*e.g.*, bio-diene), *e.g.*, 1,3-butadiene *(e.g.*, bio-1,3-butadiene), production for a particular alkenol (*e.g.*, bio-alkenol) or diol (*e.g.*, bio-diol) feedstock. In some embodiments, the rate of alkenol (*e.g.,* bio-alkenol) or diol (*e.g.*, bio-diol) feedstock passage is about 0.3 to about 10 mass units of feedstock per mass unit of catalyst/h (*i.e.*, 0.3 to 10 WHSV), or about 1.2 to about 2.0 WHSV, or may be greater than about 1 and less than about 6 WHSV in some embodiments. In some embodiments, water content in the alkenol (*e.g.*, bio-alkenol) or diol (*e.g.*, bio-diol) feedstock in contact with the catalyst in the reactor **403** (water in the alcohol (alkenol or diol) feed entering the reactor, and optionally additional water added to the feed and/or added separately into the reactor) is about 1 ppm *(i.e.*, 0.0001 wt-%) to about 93 wt-%. The output from apparatus **403** is fed to a cooling/condenser system **404**. The output

from **404** is transferred to a decanter where an aqueous solution **407** separates from the diene product **405**. The isolated water product **407** (after decanting or distillation to create the isolated alkene **406**) may be optionally passed through a filter material, and/or one or more other unit operations **408** to remove traces of organic acids or other unwanted byproducts that may be produced during the dehydration reaction, thereby producing a purified water stream **402**. Optionally, the water stream **402** may be added to the alkenol or diol feed in **401**.

[137] In some embodiments, the alcohols (*e.g.*, bio-alcohols) (alkenols (*e.g.*, bio-alkenols) or diols (*e.g.*, bio-diols)) that may be selectively dehydrated to produce dienes (*e.g.*, bio-dienes) with the catalyst compositions and methods disclosed herein may include at least one additional organic functional group. Non-limiting examples of such organic functional groups include, but are not limited to, aromatic (*e.g.*, with an aromatic ring that consists of carbon atoms or contains a mixture of carbon and heteroatoms), nitrile, alkene, or ketone functional groups. In some embodiments, the alcohol is a terminal alcohol. In other embodiments, the alcohol is not a terminal alcohol (*e.g.*, 2,3-butanediol).

[138] In some embodiments, diols are used in the dehydration methods disclosed herein. Nonlimiting examples include 1,3-butanediol, 1,4-butanediol, 2,3-butanediol, 1,4-pentanediol, 1,3-pentanediol, 2,4-pentanediol, 1,6-hexanediol, 1,5-hexandiol, 2,4-hexanediol, 1,3-hexanediol. In one embodiment, an aqueous solution of 1,3-butanediol (*e.g.*, containing from about 5 to about 95 wt-% of the diol) is dehydrated in a double dehydration to afford 1,3-butadiene, shown schematically in Fig. 14.

[139] In some embodiments, alkenols are used in the dehydration methods disclosed herein. A nonlimiting example is 2-butene-1-ol. In some embodiments of the methods disclosed herein, the 1-alkenol is an alcohol of general formula $R^1R^2CH{=}CHCH_2OH$, where R^1 and R^2 are independently H or a C_1 to C_{20} alkyl radical, or contain one or more aromatic rings containing exclusively carbon or a mixture of carbon and heteroatoms, or a nitrile, or an alkene, or a carboxylic acid, or ketone functional group, or a combination thereof.

[140] In some embodiments, the alkenol or diol may be a C_3 to C_{12} alcohol, *e.g.*, 1,12-decanediol.

[141] The diene produced in some embodiments of the methods disclosed herein may be 1,3-butadiene (*e.g.*, bio-1,3-butadiene).

[142] The input steam may be useful in double dehydration reactions that form a terminal olefin that is in conjugation with a second olefin bond (e.g. 1, 3-butadiene). Conjugated

dienes are generally more reactive and thus less stable. Dilution with solvent is commonly used to slow their decomposition/polymerization chemistry.

[143] In some embodiments of the dehydration methods disclosed herein, the alkenol or diol and water are heated to about 200 °C to 400 °C and then contacted with the dehydration catalyst. In some embodiments, the alkenol is crotyl alcohol. In one embodiment, the catalyst is "**Catalyst 2**," described *infra* in Example 8. In one embodiment, the catalyst is "**Catalyst 2**," described *infra* in Example 8, and the alkenol is crotyl alcohol. In one embodiment, the catalyst is "**Catalyst 4**," described *infra* in Example 10. In one embodiment, the catalyst is "**Catalyst 4**," described *infra* in Example 10, and the alkenol is crotyl alcohol. In one embodiment, the catalyst is "**Catalyst 5**," described *infra* in Example 12. In one embodiment, the catalyst is "**Catalyst 5**," described *infra* in Example 12, and the alkenol is crotyl alcohol. The exiting product stream may be at a temperature of about 150 °C to about 350 °C. In embodiments of the dehydration methods disclosed herein, the final product stream may contain about 10 mol% alkenol or diol or less (based on alkenol or diol fed; corresponding to about 90 mol%, or greater, alkenol or diol conversion), about 5 mol% alkenol or diol or less (based on alkenol or diol fed; corresponding to about 95mol%, or greater, alkenol or diol conversion), or less than about 0.5 mol% alkenol or diol (based on alkenol or diol fed; corresponding o about 99.5 mol%, or greater, alkenol or diol conversion). Embodiments of the methods disclosed herein may produce the diene product with about 90% to about 98% or greater selectivity for the diene.

[144] The alkenol or diol dehydration methods disclosed herein may be operated in either isothermal or adiabatic configurations.

[145] In a further embodiment, the dehydration method may include at least two reactors in sequence, with the same, similar, or better performance and yield as the single reactor system. Having the catalyst in more than one reactor can eliminate the need for external heating of the commercial reactors. Heating large commercial reactors in an isothermal manner can be cost prohibitive. In some embodiments, the product stream from the catalytic dehydration reaction (*e.g.*, **403**) is then heated again to about 200 °C to about 400 °C and contacted with a second reaction vessel containing dehydration catalyst. An example of a two-reactor process is shown schematically in FIG 5. The alkenol or diol feed **500** is added to the vaporization/mixing vessel **501** and a water feed is also added at some preselected amount. The output of **501** is transferred to the first catalyst vessel **503** which contains the dehydration catalyst (*e.g.*, **206** or **304**, prepared as described above). The output from **503** is reheated in inter-stage heater **504** and fed to a second catalyst vessel **505**.

Vessel **503** and **505** typically do not have an external source of heat applied. The product stream from **505** is taken to a cooling/condenser system **506**. The output from **506** is transferred to a decanter where an aqueous solution **508** separates from the diene product **507**. The product diene **509** may be removed, and the aqueous solution **508** can be passed through a filter and/or one or more other unit operations **510** to remove traces of organic acids or other unwanted byproducts that can be produced during the dehydration reaction, thereby producing a purified water stream **502**. Optionally, the water stream **502** may be added to the alkenol or diol feed in **501**. For those skilled in the art, this process is commonly denoted as an "adiabatic reactor" and, in some embodiments, provides the most economical and energy efficient means of dehydrating a bio-alkenol. In calculating a WHSV, the weight of alcohol feed per hour is divided by the weight of catalyst contained in both reactors. Thus in the system described above with two reactors, each separate reactor will appear to have a WHSV equaling twice that of the overall process WHSV (feed to process/total catalyst, where total catalyst is reactor 1 + reactor 2; assuming, for the explanatory case described here, that each of reactor 1 and reactor 2 is charged with the same amount of catalyst).

[146] In some embodiments of the methods disclosed herein, a device for separation of the diene product from water (e.g., an aqueous solution that contains water and other components, for example, trace organic acids and/or other byproducts) is provided. For example, water may be separated from the diene product stream by fractionation through a distillation column. In some embodiments distillation may include injecting nitrogen, steam, or other volatile inert (*e.g.*, steam-stripping of the product stream as part of the separation process). In some embodiments where the product stream includes 1,3-butadiene, the diene is recovered using methods that include steam- or nitrogen-stripping of the product stream. Alternatively, physical separation of layers can be used under conditions where the diene is a liquid, for example, when the diene product contains a sufficiently high boiling point, for example, boiling at a temperature above about 30°C. In this case, separation may optionally be achieved with a decanter (**405/407** or **507/508**, as shown schematically in FIG 4 or FIG 5, respectively). The diene product will typically be the top layer formed upon condensation and is typically siphoned from the top of the decanter vessel with the separated aqueous layer (*i.e.*, typically bottom layer) drained (*e.g.*, continuously drained) from the bottom layer (*e.g.*, **407**, **508**). The separated aqueous layer (e.g., **407**, **508**) can optionally be filtered to remove impurities and the water recycled for addition to the alkenol or diol feed. Alternatively, the diene may be recovered by liquid-

liquid extraction techniques known in the art. In some embodiments where the product stream includes 1,3-butadiene, the diene is recovered using methods that include liquid-liquid extraction.

[147] In some embodiments, the alkenol or diol/input steam is introduced to the catalyst reactor (*e.g.*, **503** or **403**) at about 1 psig to about 200 psig, about 5 psig to about 150 psig, or about 15 psig to about 100 psig. In certain embodiments, the exit pressure from the catalyst reactor(s) can be controlled by at least one back pressure regulator located on the exit side of the reactor. An increase in pressure can be useful although in some embodiments, this does not lead to an increase in the rate of dehydration (*e.g.*, an increase in WHSV at a given temperature).

[148] Although not wishing to be bound by theory, it is well known that under certain conditions, crotyl alcohol may undergo isomerization to form 3-buten-2-ol [*see, e.g.*, U.S. Patent No. 2,373,956]. In catalytic dehydration of crotyl alcohol, isomerization of crotyl alcohol may be concurrent with dehydration. Under conditions of contact with the dehydration catalyst, isomerization of crotyl alcohol to 3-buten-2-ol may occur, in which case some of the diene product will be a result of the direct dehydration of 3-buten-2-ol, in the case that the dehydration catalyst is competent for 3-buten-2-ol dehydration to diene(s). These reaction pathways are shown schematically in FIG 6.

[149] In some embodiments a diene product produced as described herein can be utilized to make a drop-in diesel or jet fuel (*e.g.*, biofuel) or a lubricant (*e.g.*, biolubricant) in an energy-efficient manner. The term "drop-in" is used to indicate the new fuel prepared from the diene product can be used in commercial or military applications without the need for equipment changes or modifications. For example, the new fuels can meet the requirements of ASTM D1655 (via ASTM D7566), the standard for turbine fuels. Thus, the new fuels can be fully compatible with existing petroleum fuel standards, be used in commercial aircraft, and therefore be considered "drop-in fuels."

[150] In one embodiment, the methods taught herein may be utilized for conversion of an alkenol (*e.g.*, crotyl alcohol) or a diol that is produced biologically, for example, in a microbial fermentation process (*e.g.*, bio-crotyl alcohol), containing small to large amounts of water, for example, water from the fermentation broth and optionally additional added water, to a diene (*e.g.*, 1,3-butadiene) with high selectivity and chemical conversion. The 1,3-butadiene in turn is useful in preparing products that are environmentally beneficial and reduce greenhouse gases compared to petroleum equivalents by virtue of the biological

production of the alcohol and an energy efficient process of making the downstream products.

[151] An example of a method for making 6-carbon monomers from a four-carbon alkenol or diol as the starting material is shown schematically in FIG 7. The alkenol or diol feed **700** is added to the vaporization/mixing vessel **701** to which water feed **702** is added at some preselected amount. The output of **701** is transferred to the catalyst vessel **703** which contains the dehydration catalyst (*e.g.*, **206** or **304**, prepared as described above). The output from **703** is fed to a cooling/condenser system (**704**). The output from **704** is transferred to a decanter where an aqueous solution **707** separates from the diene or bio-diene product **705.** The aqueous solution **707** can be passed through a filter **708** and/or one or more unit operations to remove traces of organic acids or other unwanted byproducts that can be produced during the dehydration reaction, thereby producing a purified water stream **702**. Optionally, the water stream **702** may be added to the alcohol feed in **701**. The diene product **705** is fed to a dryer (**709**) (*e.g.*, a vessel charged with a dessicant medium, such as molecular sieves or similar inorganic drying agent) and subsequently undergoes a catalyzed double hydrocyanation to afford a product stream of adipoylnitrile (**710**). The product in **710** is fed to a hydrolysis reactor (**712**) to afford adipic acid (**713**) or reduced with hydrogen using a catalyst in reactor (**713**) to afford the products hexamethylene diamine (**714**).

[152] In embodiments of the dehydration methods disclosed herein in which the alkenol or diol is produced biologically (*e.g.*, bio-1-alkenol or bio-diol), contaminants (*e.g.*, organic contaminants) may be present in the alcohol-water mixture (*e.g.*, from fermentation broth), including but not limited to, esters, aldehydes, and/or ketones. Such contaminants can be tolerated in the dehydration reactions disclosed herein at various levels depending upon the alkenol (*e.g.*, bio-alkenol) or diol (*e.g.*, bio-diol) feedstock used. In some embodiments, up to about 0.5 wt-%, or up to about 5 wt-% contaminants (*e.g.*, organic contaminants) may be tolerated, for example, when crotyl alcohol (e.g., bio-crotyl alcohol) is the starting material.

[153] In some embodiments of the methods disclosed herein, removal of final traces of water and oxygenated organics (*e.g.*, ethers, aldehydes, and/or aldol-type products) from the diene (*e.g.*, bio-diene) product can be accomplished by passing the diene stream through a fixed bed of separation media, for example, activated alumina, molecular sieves, Celite, activated charcoal, size-exclusion type of media (*e.g.*, selective gas permeation membranes), or a combination thereof. For example, the media may be contained in a vessel shown schematically in FIG 7 as dryer **709**. In some embodiments, reactivation of the fixed-bed materials may be possible by heating under a purge of hydrocarbon or inert gas while

heating to temperature of about 50° to about 200° C, or about 100° C. In one embodiment, a series of membranes and/or bed of ionic-liquids that are well known for permitting selective passage of non-polar molecules and retaining more polar organics (*e.g.*, ethers and alcohols) can be used to dewater the diene product.

Biological production of alkenols and diols

[154] In some embodiments, alkenols and/or diols may be produced via microbial fermentation. The alkenol and/or diol products may be catalytically converted to diene products as described herein. In some embodiments, alkenol and/or diol products may be at least partially separated from the fermentation medium and dehydrated to produce diene products using catalyst compositions as described herein.

[155] In some embodiments, a microorganism that is used for production of alkenol(s) and/or diol(s) has been modified to produce a greater amount of the alkenol(s)and/or diol(s) than the parent microorganism from which it is derived. In other embodiments, a microorganism is used that produces the alkenol(s) and/or diol(s) via existing (*e.g.*, native) enzyme(s) and/or biosynthetic pathways. In some embodiments, the microorganism is a *Clostridium* strain.

[156] In some embodiments, microbial production of crotyl alcohol is achieved through the ability of the native promiscuous alcohol dehydrogenase (adhE, adhe2) in a microorganism, *e.g.*, a *Clostridium* strain, to convert crotonyl-CoA to crotonaldehyde. In some embodiments, microbial production of crotyl alcohol is achieved through the ability of the native promiscuous alcohol dehydrogenases (adhE, adhE2, bdhAB) to convert the crotonaldehyde to crotyl alcohol.

[157] In some embodiments, microbial production of crotyl alcohol is achieved through the deletion or inactivation of the butyryl-CoA dehydrogenase (bcd) and/or crotonyl-CoA reductase gene, or similar genes that are a part of butanoate/butanol, fatty acid, and amino acid degradation pathways in a microorganism., *e.g.*, a *Clostridium* organism.

[158] In some embodiments, the production of crotyl alcohol from an engineered microorganism, *e.g,* a *Clostridium* microorganism, is enhanced through treatment of the engineered *Clostridium* (*e.g.*, engineered to delete or inactivate the bcd gene) with one or more chemical mutagen(s), followed by selection under that bias the selection towards strains with increased crotyl alcohol production, selection in the presence of crotyl alcohol.

[159] In some embodiments, the production of crotyl alcohol from a non-engineered or non-modified microorganism, *e.g.*, a *Clostridium* microorganism, is enhanced by treatment

with one or more chemical mutagen(s), followed by selection under conditions that bias the selection towards strains with increased crotyl alcohol production, for example, but not limited to, selection in the presence of crotyl alcohol and/or another related alcohol, *e.g.*, propanol, pentenol, pentanol.

[160] A strategy is described herein whereby a microorganism, *e.g.*, a *Clostridium* microorganism, that produces *n*-butanol is converted into a microorganism that produces crotyl alcohol, for example, by deletion, inhibition, and/or inactivation of the activity of one or more enzyme(s) (*e.g.*, bcd, phosphotransbutyrylase (ptb), butyrate kinase (bk), CoT (CoA transferase), acetoacetate decarboxylase (aad)). In some embodiments, deletion, inhibition, and/or inactivation of CoT and/or aad reduces or eliminates production of acetone in an ABE producing microorganism, *e.g.*, a *Clostridium* microorganism, which improves crotyl alcohol yield. Other strategies include converting a microorganism, e.g., a *Clostridium* microorganism, into a microorganism that produces crotyl alcohol by overexpression of one or more enzyme(s) (*e.g.*, an enzyme of a glycolytic or pentose phosphate pathway that converts glucose and/or xylose to acetyl-CoA, a thiolase enzyme that converts acetyl-CoA to acaetoacetyl-CoA, a 3-hydroxy butyryl CoA dehydrogenase enzyme that converts acetoacetyl-CoA to 3-hydroxybutyryl-CoA, a crotonase enzyme that converts 3-hydroxybutyryl-CoA to crotonyl-CoA, an aldehyde dehydrogenase enzyme that converts crotonyl-CoA to crotonaldehyde, an alcohol dehydrogenase enzyme that converts crotonaldehyde to crotyl alcohol). In some embodiments, at least one enzyme of a butanol/butanoate pathway (*e.g.,* bcd) is deleted, inhibited, or inactivated, and at least one enzyme of a crotyl alcohol pathway (*e.g.*, an alcohol dehydrogenase enzyme) is overexpressed.

[161] Other strategies are described herein in which non-engineered microorganisms (e.g., microorganisms that produce *n*-butanol) may produce crotyl alcohol by adjustment of culture conditions to favor production of this product. For example, one or more substance(s), including but not limited to, one or more divalent metal ion(s) (e.g., Ca^{2+}, Co^{2+}, Zn^{2+}, and/or Mn^{2+}), iodoacetamide, myristoyl-CoA, N-ethylmaleimide, and/or palmitoyl-CoA may be included in the growth medium, thereby increasing production of crotyl alcohol versus a growth medium that does not include the substance(s). Such substance(s) may serve as inhibitors of one or more enzyme(s), including but not limited to, bcd, thereby increasing production of crotyl alcohol by the microorganism. In some embodiments, an SiHybrid may downregulate the activity of one or more enzyme(s), such as bcd.

[162] Methods are provided for producing one or more alkenol(s) or diol(s) in a microbial fermentation. The methods include culturing a microorganism that produces the alkenol(s) and/or diol(s) of interest (*e.g.*, crotyl alcohol) in a medium that contains a carbon source, such as, but not limited to, soluble sugar molecules produced from biomass (*e.g.*, in an acid and/or enzymatic hydrolysate of biomass) to support microbial growth for production of the alkenol(s) and/or diol(s).

[163] The methods for production of alkenol(s) and/or diol(s) (*e.g.*, crotyl alcohol) herein include fermentation with an engineered, mutagenized, or non-engineered, non-mutagenized microorganism (*e.g.*, *Clostridium* microorganism) as described herein in a bioreactor in a growth medium that contains a carbon source, such as, but not limited to, a liquid sugar-containing extract from biomass, such as a hydrolysate or conditioned hydrolysate (*e.g.*, conditioned to remove at least a portion of one or more inhibitor(s) of microbial growth and/or bioproduct (*e.g.*, alkenol and/or diol) production), prepared from biomass (*e.g.*, lignocellulosic biomass). In some embodiments, a liquid sugar containing extract such as cane juice and/or molasses is included in the growth medium.

[164] In some embodiments, the bioproduct production includes fermentation with a microorganism as described herein in an immobilized cell bioreactor (*i.e.*, a bioreactor containing cells that are immobilized on a support, *e.g.*, a solid support). In some embodiments, an immobilized cell bioreactor provides higher productivity due to the accumulation of increased productive cell mass within the bioreactor compared with a stirred tank (suspended cell) bioreactor. In some embodiments, the microbial cells form a biofilm on the support and/or between support particles in the growth medium.

[165] In other embodiments, for example but not limited to, embodiments in which a hydrolysate composition containing both liquid hydrolysate and solid residues is used, microorganisms may be grown in a non-immobilized system, such as an agitated fermentation reactor, *e.g.*, designed to provide adequate conditions for fermentation, including but not limited to mixing of components, gas removal, temperature control, and/or the ability to add and/or remove material from the reactor. Several fermentation operational moieties exist, including but not limited to batch, fed-batch, and continuous in single or multiple reactor configurations. Exemplar reactor types include but are not limited to agitated tanks, *e.g.*, where agitation is effected by a mechanical impeller, the addition and withdrawal of material, the addition of gas, and/or the recirculation of fermentation gas; corn and/or cane ethanol fermentation tanks; pharmaceutical fermentation vessels; vacuum fermentation systems; air-lift type reactors; fluidized bed reactors; anaerobic digestors; and

activated sludge reactors. In some embodiments, an extractive fermentation process is used (*e.g.*, gas stripping, liquid extraction, vacuum fermentation, extraction by absorption and/or adsorption by a solid material such as a polymeric material).

[166] In some embodiments, the alkenol and/or diol (*e.g.*, crotyl alcohol) production process herein includes continuous fermentation of a microorganism (continuous addition of conditioned hydrolyzed feedstock and withdrawal of product stream). Continuous fermentation minimizes the unproductive portions of the fermentation cycle, such as lag, growth, and turnaround time, thereby reducing capital cost, and reduces the number of inoculation events, thus minimizing operational costs and risk associated with human and process error.

[167] Fermentation may be aerobic or anaerobic, depending on the requirements of the bioproduct-producing microorganism.

[168] As known in the art, in addition to an appropriate carbon source, fermentation media must contain suitable nitrogen source(s), mineral salts, cofactors, buffers, and other components suitable for the growth of the cultures and promotion of the enzymatic pathway necessary for the production of the desired bioproduct. In some embodiments, salts and/or vitamin B12 or precursors thereof are included in the fermentation media. In some cases, hydrolyzed biomass may contain some or all of the nutrients required for growth, minimizing or obviating the need for additional supplemental material.

[169] One or more microorganism that is capable of producing one or more alkenol(s) or diol(s) of interest (*e.g.*, crotyl alcohol) may used in the fermentation methods described herein. In embodiments in which two or more microorganisms are used, the microorganisms may be the same or different microbial species and/or different strains of the same species.

[170] In some embodiments, the microorganisms are bacteria or fungi. In some embodiments, the microorganisms are a single species. In some embodiments, the microorganisms are a mixed culture of strains from the same species. In some embodiments, the microorganisms are a mixed culture of different species. In some embodiments, the microorganisms are an environmental isolate or strain derived therefrom.

[171] The following examples are intended to illustrate, but not limit, the invention.

EXAMPLES

Example 1

Testing the effect of crotyl alcohol on *Clostridium n*-butanol fermentations

[172] The effect of crotyl alcohol in the growth medium of a *Clostridium* fermentation was investigated. Production of *n*-butanol by a butanol-producing *Clostridium* strain was determined at various concentrations of crotyl alcohol in the fermentation medium. Seed culture and growth media were J2 for JR22. J2 media: A liquid yeast based media containing 6% glucose as a carbohydrate source. Crotyl alcohol was added to the J2 media to the following final concentrations: 0, 0.1, 1,2,4,6, and 8g/L. Crotyl alcohol concentration in the culture media was confirmed by submitting samples for HPLC analysis prior to inoculation. All cultures were inoculated at 10% volume from a seed cultured in J2 with 0 g/L crotyl alcohol. Culture volumes were 4 mL, grown in 10 mL conical tubes. Media and culture vials were deoxygenated prior to growth in an anaerobic chamber. Typically small scale cultures are grown in 15 mL conical tubes, where there are screw thread caps in place to reduce butanol evaporation but pressure from gas generated during the fermentation can be released. Conical tubes were used to reduce the possibility of evaporation of the crotyl alcohol during the fermentation. Time points were taken at 24 and 48 h, a single time point per vial. At each time point the absorbance at 600 nm was measured and a sample was submitted for HPLC analysis. Results for crotyl alcohol addition are shown as a percent of the control (no crotyl alcohol addition) in Fig. 15.

[173] Butanol tolerance of JR22 was investigated, for comparison with crotyl alcohol tolerance. The seed culture and growth media used were molasses media. (Molasses media: A liquid yeast based media containing 6% molasses as a carbohydrate source). Butanol was added to the molasses media to the following final concentrations: 0, 3, 4.5, 6.5, 8, 10, 12, 13, and 15 g/L. Butanol concentration in the culture media was confirmed by submitting samples for HPLC analysis prior to inoculation. All cultures were inoculated at 10% volume from a seed cultured in molasses media with 0 g/L butanol added to the media. Cultures volumes, growth conditions and sampling were performed as described for the crotyl alcohol samples above. Results for *n*-butanol addition are shown in Fig. 16.

[174] Data showing total solvent production (butanol, acetone, and ethanol) as a function of added crotyl alcohol are shown in Fig. 17.

[175] Data showing the uptake of crotyl alcohol, accounting for the increased total solvent production above the maximum theoretical based on sugar uptake, are shown in Table 1.

TABLE 1

Crotyl alcohol added (g/L)	Crotyl alcohol consumed (g/L)
0	0
1	0.7
2	1.5
4	3.1
6	4.0
8	2.0

[176] The data above demonstrate that the enzymatic steps are indeed non-selective for the substrate. These data provide direct evidence the enzymatic steps (adhE and adh2) are promiscuous in nature.

Example 2

Testing the effect of crotonic acid on *Clostridium n*-butanol fermentations

[177] The effect of crotyl alcohol in the growth medium of a *Clostridium* fermentation was investigated. Production of *n*-butanol by *Clostridium* was determined at various concentrations of crotyl alcohol in the fermentation medium. Seed culture and growth media were J2 for JR22. J2 media: A liquid yeast based media containing 6% glucose as a carbohydrate source. Crotonic acid was added to the J2 media to the following final concentrations: 0, 3, 6 g/L. Crotonic acid concentration in the culture media was confirmed by submitting samples for HPLC analysis prior to inoculation (data shown in tables below for crotonic acid added indicates the actual amount present as measured by HPLC). All cultures were inoculated at 20% volume from a seed cultured in J2 (strain JR22) with 0 g/L crotonic acid. Culture volumes were 100 mL, grown in 250 mL shake flasks. Shake flasks were deoxygenated prior to growth in an anaerobic chamber. Typically small scale cultures are grown in 250 mL shake flasks where there are screw thread and pressure releasing valves. At each time point the pH and absorbance at 600 nm was measured and a sample was submitted for HPLC analysis.

[178] Results for crotonic acid addition demonstrates Glucose uptake over time are shown in Fig. 18. Data showing the consumption of crotonic acid as a function of time are shown in Fig. 19. Plots showing the n-butanol titer as a function of crotonic acid added are shown in Fig. 20.

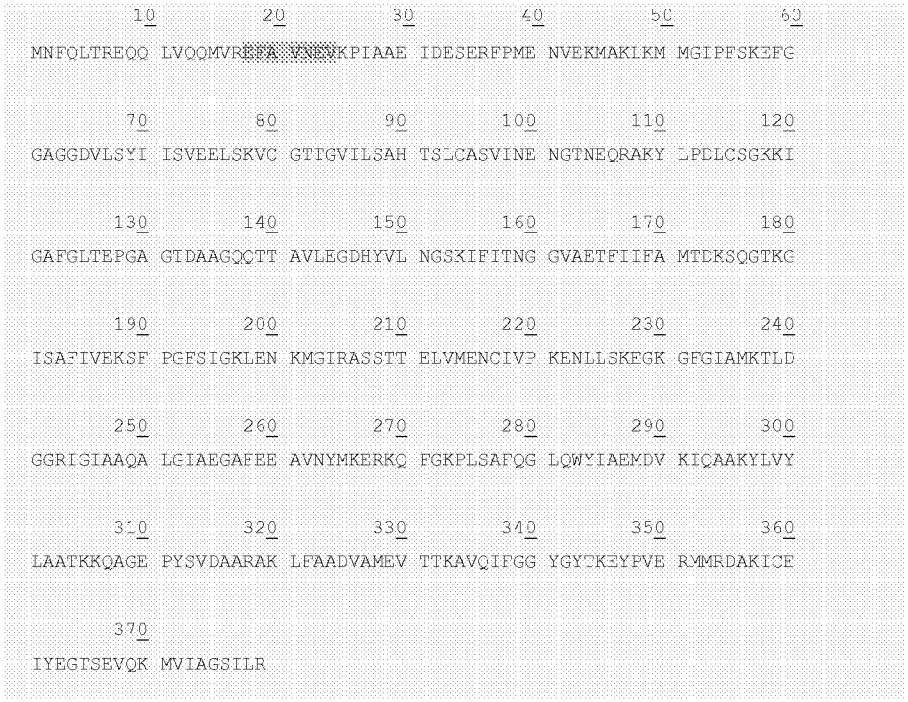
Example 3

Downregulation of the bcd gene by siHybrids in *Clostridium* for the production of crotyl alcohol from lignocellulosic hydrolysate or other fermentable feedstock with subsequent conversion of crotyl alcohol to butadiene via a dehydration catalyst

[179] This example demonstrates the engineering of a *Clostridium* strain that has the native genes shown in Fig. 21. The butanol production pathway in *Clostridium acetobutylicum* includes Ack, acetate kinase; Pta, phosphotransacetylase; AdhE, aldehyde/alcohol dehydrogenase; Thl, thiolase; Hbd, 3-hydroxybutyryl-CoA dehydrogenase; Crt, crotonase; Bcd, butyryl-CoA dehydrogenase; Ptb, phosphotransbutyrylase; Buk, butyrate kinase; CtfA/B, CoA transferase; Adc, acetoacetate decarboxylase; NADH, nicotinamide adenine dinucleotide (reduced form). The single reactions do not represent stoichiometric fermentation balances. (Scheme from Mazzoli (2012) www.ncbi.nlm.nih.gov/pmc/articles/PMC3962139/).

[180] A bcd siHybrid is designed to downregulate butyryl-CoA dehydrogenase without the need for time consuming plasmid production and transformation. With the bcd (butyryl-CoA dehydrogenase) gene silenced utilizing siHybrids to downregulate the bcd gene, the native promiscuous alcohol dehydrogenase (adhE, adhe2) can convert crotonyl-CoA to crotonaldehyde. In addition, the native promiscuous alcohol dehydrogenases (adhE, adhE2, bdhAB) can also convert the crotonaldehyde to crotyl alcohol as shown in Fig. 22. The Crotyl Alcohol Production Pathway in an engineered *Clostridium* species with inactivation of the bcd gene includes Ack, acetate kinase; Pta, phosphotransacetylase; AdhE, AdhE2, aldehyde/alcohol dehydrogenase; BdhA, BdhB, butanol dehydrogenase; Thl, thiolase; Hbd, 3-hydroxybutyryl-CoA dehydrogenase; Crt, crotonase; Bcd, butyryl-CoA dehydrogenase; Ptb, phosphotransbutyrylase; Buk, butyrate kinase; CtfA/B, CoA transferase; Adc, acetoacetate decarboxylase; NADH, nicotinamide adenine dinucleotide (reduced form). The single reactions do not represent stoichiometric fermentation balances.

First, a siHybrid is constructed using a random twenty-one base pair sequence from the published bcd gene sequence of *C. beijerinckii* (NCIMB 8052). The amino acid sequence of the bcd gene, Cbei_0322, is depicted in SEQ ID NO:1. The siHybrid is constructed with a random 7 amino acid length segment from this sequence. The region chosen for SiHybrid construction is highlighted.



(SEQ ID NO:1)

[181] Region sense and antisense strands are constructed with two nucleotide 3‘ uridine over hangs (GenScript, Piscataway, NJ). The region from amino acids 18-24 of the bcd gene, Cbei_0322 was chosen for its DNA homology to C. beijerinckii strain NRRL B593 and *C. saccharobutylicum* DSM 13864. The sequence was queried for homology with other unrelated genes using the BLAST (basic local alignment search tool) bioinformatics tool. The closest additional sequence shares only 14 of the 21 base pairs from amino acid region 18-24. Therefore, the siHybrid should not interfere with the translation of any additional genes. The sense strand is composed of RNA of the sequence 5’ CUU CAU UUA CUG CGA AUU CUC UU 3’. (SEQ ID NO:2) The antisense strand is composed of DNA of the sequence 5’ GAG AAT TCG CAG TAA ATG AAG TT 3’. (SEQ ID NO:3). Sense and antisense strands are annealed together in equimolar amounts in the presence of 10 mM Tris-HCl (pH 8.0) by denaturing for 5 minutes at 94° C and then reannealed at 53° C for 3 hours. Annealed strands are then slowly cooled to room temperature. This siHybrid is added to the fermentation medium at a concentration of 4 μg/ml.

[182] A culture of C. beijerinckii is started overnight from glycerol stock in minimal medium absent of bcd siHybrid strands. When cultures reach an optical density at 600nm of 1.0, a 10% inoculation is performed into a fermentataion medium (e.g., lignocellulosic hydrolysate), for example, with defined added nutrients under anaerobic conditions in a

bioreactor containing either 4 μg/ml of the bcd siHybrid, or medium absent of siHybrid. Cultures are incubated anaerobically, shaking at 100 rpm, at 32 C for 48 hours, with time points taken at 24, 48, and 72 hours. Fermenation broth is filtered and analyzed by HPLC for butanol, butyric acid, crotyl alcohol, and crotonic acid. If bcd activity is downregulated, HPLC results from siHybrid-containing medium will show higher amounts of crotyl alcohol and crotonic acid, while producing less butanol and butyric acid than the control. Fermenation broth is also analyzed for protein suppression (e.g., Bcd protein expression levels) by Western blot.HPLC is used to monitor the crotyl alcohol production during fermentation. Western blot analysis is also performed to evaluate the Bcd protein expression levels.

[183] At the end of fermentation, crotyl alcohol may be collected from the fermentation broth and is converted with a dehydration catalyst to bio-1,3-butadiene as described herein.

[184] The DNA sequence of Cbei_0322 from NCIMB 8052 is depicted in SEQ ID NO:4. The region chosen for SiHybrid construction is highlighted.

atgaatttccaattaactagagaacaacaattagtacaacaaatggttagagaattcgcagtaaatgaagttaagccaatagctgctga
aatcgacgaatcagaaagattccctatggaaaacgttgaaaaaatggctaagcttaaaatgatgggtatcccattttctaaagaatttgg
tggagcaggcggagatgttctttcatatataatatctgtggaagaattatcaaaagtttgtggtactacaggagttattctttcagcgcata
catcattatgtgcatcagtaattaatgaaaatggaactaacgaacaaagagcaaaatatttgccagatctttgtagtggtaagaaaatcg
gtgctttcggattaacagaaccaggcgctggtacagatgctgcaggacaacaaacaactgctgtattagaaggagaccattatgtatt
aaatggttcaaaaatcttcataacaaatggtggagttgctgaaactttcataatatttgctatgacagataagagtcaaggaacaaaagg
aatttctgcattcatagtagaaaagtcattcccaggattctcaataggaaaattagaaaacaagatggggatcagagcatcttcaacta
ctgagttagttatggaaaactgtatagtaccaaaagaaaacctacttagcaaagaaggtaagggatttggtatagcaatgaaaactctt
gatggaggaagaattggtatagctgctcaagctttaggtattgcagaaggagcttttgaagaagctgttaactatatgaaagaaagaa
aacaatttggtaaaccattatcagcattccaaggattacaatggtatatagctgaaatggatgttaaaatccaagctgctaaatacttagt
atacctagctgcaacaaagaagcaagctggtgagccttactcagtggatgctgcaagagctaaattatttgcggcagatgttgcaatg
gaagttacaactaaagcagttcaaatctttggtggatatggttacactaaggaataccagtagaaagaatgatgagagatgctaaaata
tgcgaaatctacgaaggaacttcagaagttcaaaagatggttatcgcaggaagcattttaagatag (SEQ ID NO:4)

[185] The DNA sequence of the bcd gene *from C. beijerinckii* strain NRRL B593 is depicted in SEQ ID NO:5. The region chosen for siHybrid construction is highlighted,

atgaatttccaattaactagagaacaacaattagtacaacaaatggttagagaattcgcagtaaatgaagttaagccaat
agctgctgaaatcgacgaaacagaaagattccctatggaaaacgttgaaaaaatggctaagcttaaaatgatgggtatc
ccattttctaaagaatttggtggagcaggcggagatgttctttcatatataatagctgtggaagaattatcaaaagtttgtg
gtactacaggagttattctttcagcgcatacatcattatgtgcatcagtaattaatgaaaatggaactaacgaacaaagag
caaaatatttacctgatctttgcagcggtaaaaagatcggtgctttcggattaactgaaccaggtgctggtacagatgctgc
aggacaacaaacaactgctgtattagaaggggatcattatgtattaaatggttcaaaaatcttcataacaaatggtggagt
tgctgaaactttcataatatttgctatgacagataagagtcaaggaacaaaagaatttctgcattcatagtagaaaagtca
ttcccaggattctcaataggaaaattagaaaataagatggggatcagagcatcttcaactactgagttagttatggaaaac
tgcatagtaccaaaagaaaacctacttagcaaagaaggtaagggatttggtatagcaatgaaaactcttgatggaggaa
gaattggtatagctgctcaagctttaggtattgcagaaggagcttttgaagaagctgttaactatatgaaagaaagaaaac
aatttggtaaaccattatcagcattccaaggattacaatggtatatagctgaaatggatgttaaaatccaagctgctaaata
cttagtatacctagctgcaacaaagaagcaagctggtgagccttactcagtagatgctgcaagagctaaattatttgctgc
agatgttgcaatggaagttacaactaaagcagttcaaatctttggtggatatggttacactaaagaatacccagtagaaag
aatgatgagagatgctaaaatatgcgaaatctacgaaggaacttcagaagttcaaaagatggttatcgcaggaagcattt
taagata

(SEQ ID NO:5)

[186] The DNA sequence of the bcd gene from *C. saccharobutylicum* DSM 13864 is depticted in SEQ ID NO:6. The region chosen for siHybrid construction is highlighted.

atgaattttaagttaactagagaacaagaattaatacaacaaatggttagagaattcgcagtaaatgaagttaaaccaat
agctgctgaaatcgatgagacagaacaatttccgatggaaaatgttaaaaaaatggctgaattaggaatgatgggtattc
cattttctaaagaagtaggaggagcaggcggagatgttctttcttatataatagctgtagaagaattatcaaaagtgtgtg
gtactacaggtgttatactttcagcacatacatcattatgtgcatcagtaattaacgaaaatggtactccagctcaaaaaga
aaaatatttagcagatctttgtagtggtaagaaaattggtgctttggtttaactgaaccaggtgctggtacagatgctgctg
gacaacaaacaacagcaacattagaaggagatcattatgtattaaatgggtcaaaaatattcataacaaatggtggagtt
gctgaaactttcataattttgctatgacagataagagccaaggaactagaggaatttctgcatttgtagtagaaaaatcat
tccctggattctcagtaggaaaattagaaaacaagatgggtattagagcatcttcaactactgaattagttatggaaaact
gtatagtaccaaaagaaaacttacttggtaaagaaggtaagggatttggtatagcaatgaagactcttgatggaggaag
aattggtatagctgctcaagctttaggtattgcagaaggagcttttgaagaagcagttgcatacatgaaagaaagaaaac
aatttaataaaccactttcagcattccaaggattacaatggtatattgctgaaatggacgttaagatagacgctgctaagc
atttagtatataaagcagcatgcaagaaacaagcgggtcaaccttattcagttgatgcggctagagctaagttgtttgcag
cagacgttgcaatggaagttacaactaaggcagtacaaatctttggtggatatggttacactaaggaatacccagtagaa
agaatgatgagagatgctaagataactgaaatctatgaaggaacttcagaagttcaaaagatggttattgcaggaagcat
tttaagatagg

(SEQ ID NO:6)

Example 4

Engineered *Clostridium* via bcd inactivation for the production of crotyl alcohol from lignocellulosic hydrolysate or other fermentable feedstock with subsequent conversion of crotyl alcohol to butadiene via a dehydration catalyst

[187] Example 3 describes the engineering of a *Clostridium* strain that has the native genes with the bcd (butyryl-CoA dehydrogenase) gene inactivated via an intron. The native promiscuous alcohol dehydrogenase (adhE, adhe2) can convert crotonyl-CoA to

crotonaldehyde. In addition, the native promiscuous alcohol dehydrogenases (adhE, adhE2, bdhAB) can also convert the crotonaldehyde to crotyl alcohol.

[188] Preliminary inactivation of bcd is done using targetron, a gene knockout system with proven efficacy in *Clostridium* species (available from Sigma Aldrich). The targetron system works by inserting a group II intron via the activity of an RNA-protein complex into the gene of interest. Primers are generated for specific targeting of the group II intron into the bcd gene. The re-targeted intron is generated by PCR, and this segment of the group II intron is ligated into a linearized vector that contains the rest of the group II intron components, as well as an antibiotic resistance gene that is flanked by Flp recombinase recognition sites. The vector is transformed by methods known in the art into the *Clostridium* host and the RNA-protein complex is expressed. This RNA-protein complex locates the genomic target, inserts RNA and reverse transcribes cDNA. Host enzymes repair and create a permanent insertion into the bcd gene of the group II intron. Chromosomal insertion mutants are selected with the antibiotic marker. The antibiotic resistance marker is removed by transformation with a vector that expresses Flp, excising the antibiotic resistance marker. Verification of disruption of the bcd gene is performed by colony PCR using gene and intron specific primers.

[189] Gene disruption using targetron is performed with pJIR750ai containing a group II intron that has been retargeted to the bcd gene in *Clostridium beijerinckii*, as shown in Fig. 23. The intron is expressed from a beta-2-p promoter. This intron contains a Kanamycin resistance (KanR) marker with its promoter (P1) inserted in the orientation opposite that of the intron transcription. This KanR marker is disrupted by a self-splicing td group I intron inserted in the forward orientation. During retrotransposition via an RNA intermediate, the td intron is spliced, activating the KanR marker, which is then selected after the intron has integrated into the DNA target site. Flp recombinase recognition (FRT) sites flank the KanR marker gene. Expression of the Flp protein from a separate plasmid leads to efficient excision of the KanR marker from the integrated targetron, enabling multiple sequential disruptions.

[190] The engineered *Clostridium* strain is cultivated from a glycerol stock and inoculated into a lignocellulosic hydrolysate or other growth medium with nutrients for growth of the microorganism under anaerobic conditions in a bioreactor. HPLC is used to monitor the crotyl alcohol production during fermentation. At the end of fermentation, crotyl alcohol in the fermentation broth, optionally separated from the fermentation broth, is converted to 1,3-butadiene with a dehydration catalyst as disclosed herein

Example 5

Inactivation of bcd gene by homologous recombination to insert a selectable marker or deletion of a portion of the bcd gene

[191] The bcd gene and approximately 1kb of flanking regions (sequence homology upstream and downstream) is cloned into a suicide vector. A selectable marker, such as erythromycin resistance, is used to replace all or a portion of the bcd gene while preserving the integrity of the operon with an in frame deletion. The plasmid is transformed into *Clostridium* by methods known in the art. Since the plasmid is unable to replicate in *Clostridium*, *Clostridium* with resistance to the selectable marker undergoes homologous recombination at the upstream, downstream, or at both upstream and downstream sequences. Selectable marker resistant transformants are screened by PCR to identify double recombinant events. The selectable marker maybe removed using strategies known in the art to create a markerless knockout.

[192] The resulting engineered strain has an inactivated bcd gene and is able to produce crotyl alcohol. The engineered *Clostridium* strain is cultivated from a glycerol stock and inoculated into a lignocellulosic hydrolysate or other growth medium with nutrients for growth of the microorganism under anaerobic conditions in a bioreactor. HPLC can be used to monitor the crotyl alcohol production during fermentation. At the end of fermentation, crotyl alcohol in the fermentation broth, optionally separated from the fermentation broth, is converted to 1,3-butadiene with a dehydration catalyst as disclosed herein..

Example 6

Engineered and optimized *Clostridium* via bcd inactivation for production of crotyl alcohol, followed by directed evolution to optimize crotyl alcohol production from lignocellulosic hydrolysate or other fermentable feedstock with subsequent conversion of crotyl alcohol to butadiene via a dehydration catalyst

[193] Methods described in Example 4 will be followed to engineer *Clostridium* with an inactivated bcd gene. After inactivation of the bcd gene the production of crotyl alcohol will be optimized via directed evolution. The bcd knockout *Clostridium* is treated with mutagen (*e.g.*, both ethyl methanesulfonate (EMS) and N-methyl-N`-nitro-N-nitrosoguanidine (NTG) have proven efficacy for mutagenesis in *Clostridium*). Mutagenized bacteria are then grown in the presence of selection to bias the bacterial pool toward the desired outcome. Growth in the presence of crotyl alcohol is employed to select

organisms with increased tolerance to crotyl alcohol. Growth in the presence of lignocellulosic compounds typically found to be inhibitory may also be employed, to select for increased tolerance to these compounds found in hydrolysates containing lignin. Selections for increased productivity may also be employed.

[194] In addition or alternatively, metabolic flux analysis (MFA) is used for the determination of metabolic pathway fluxes in order to 1) verify that deletion of bcd leads to a significant accumulation of crotyl alcohol, 2) identify nodes that are potential bottlenecks to crotyl alcohol biosynthesis, and 3) identify competing biochemical pathways that direct carbon flow away from crotyl alcohol for further engineering. In this approach, the intracellular fluxes are calculated by using a stoichiometric model constructed from major pathway reactions and mass balances around intracellular metabolites. Intracellular fluxes are estimated based on measurable quantities, in particular, the rate of formation of extracellular metabolites. A set of measured extracellular fluxes including uptake rates of substrates and secretion rates of metabolites are used as inputs. The resulting flux calculation is a metabolic flux map showing biochemical reactions along with an estimate of the steady state rate at which each reaction occurs. This information is used to further engineer and optimize the strain for crotyl alcohol production.

[195] The resulting engineered *Clostridium* strain optimized for crotyl alcohol tolerance is cultivated from a glycerol stock and inoculated into a growth medium under anaerobic conditions in a bioreactor. HPLC can be used to monitor the crotyl alcohol production during fermentation. At the end of fermentation, crotyl alcohol in the fermentation broth, optionally separated from the fermentation broth, is converted to 1,3-butadiene with a dehydration catalyst as disclosed herein.

[196] The γ-alumina used for preparation of Catalysts **1** – **5** (Examples 7 – 10, 12, *infra*) was supplied by Strem Chemicals (min. 97%; surface area 210 m^2/g; product number 13-2525; lot number 23996900).

Example 7

[197] **Catalyst 1** was made by scaling the following method sufficient to prepare about 50g of finished catalyst: 1.5 wt-% of NaOH was loaded onto γ-alumina by incipient wetness impregnation. The NaOH solution contained 0.06 g of NaOH dissolved in water to make 4

mL of solution. The clear solution was added to 4 g of γ-alumina and mixed for about 5 min. The impregnated carrier was then dried at 200°C for 16 h to obtain the final catalyst.

Example 8

[198] **Catalyst 2**. γ-Alumina (60 g) was slurried in water (a portion, sufficient to create a slurry, taken from a total of 100 mL); $NaHSO_4$ (5 g) was dissolved in the remainder of the total of 100 mL of water to give a clear solution; the alumina slurry and $NaHSO_4$ solution were combined and stirred well; and the resulting mixture was taken to free-flowing dryness, by drying (ambient to about 200°C) overnight under a blanket of dry nitrogen with a slow nitrogen purge.

Example 9

[199] **Catalyst 3** was made by loading 5.0g $Ca(H_2PO_4)_2 \cdot H_2O$ onto 45g γ-alumina by incipient wetness impregnation. The impregnated carrier was then calcined at 300°C for 24 h to obtain the final catalyst.

Example 10

[200] **Catalyst 4**. γ-Alumina (49 g) was slurried in 40 mL of water; $Ca(H_2PO_4)_2 \cdot H_2O$ (4.30 g, 17.1 mmol) and H_3PO_4 (0.5 mL of 85%, 0.425 g of H_3PO_4, 4.34 mmol) were dissolved in 50 mL of water to give a clear solution; the alumina slurry and $Ca(H_2PO_4)_2/H_3PO_4$ solution were combined and stirred well; and the resulting mixture was taken to free-flowing dryness, by drying (ambient to about 200°C) overnight under a blanket of dry nitrogen with a slow nitrogen purge.

Example 11

Catalytic performance in dehydration of crotyl alcohol to 1,3-butadiene.

[201] Catalyst examples **1-4** prepared above were tested in a fixed-bed test unit. In the test, a crotyl alcohol feed (either neat, or diluted as indicated in Table 2, below) was passed over a catalyst bed (42 – 54g) at 1 atm and a WHSV ([g crotyl alcohol]/[g catalyst]/hour) and bed temperature, as indicated in Table 2, below. The reaction products at the reactor outlet were trapped in serial traps, the first, initially empty, and maintained at 0 - 5°C, the second, initially toluene-filled, and maintained at -78°C, and then sampled and analyzed offline by gas chromatography with flame ionization detection.

TABLE 2. Crotyl Alcohol Dehydration Results.

	WHSV (h^{-1})	T (°C)	Diluent (wt%)	Crotyl Alcohol conversion	1,3-butadiene selectivity
Catalyst **1**	0.2	380	none	90%	<40%
Catalyst **2**	0.8	280	water (10)	99%	90%
Catalyst **3**	0.3	380	none	99%	<50%
Catalyst **4**	0.9	280	water (10)	99%	95

Example 12

[202] **Catalyst 5**. γ-Alumina (80 g) was slurried in 70 mL of water; $Ca(H_2PO_4)_2 \cdot H_2O$ (6.8 g, 27.0 mmol) was dissolved in 50 mL of water; H_3PO_4 (0.82 mL of 85%, 0.694 g of H_3PO_4, 7.09 mmol) was diluted in 10 mL of water; the $Ca(H_2PO_4)_2$ and H_3PO_4 solutions were combined to give a clear solution; the alumina slurry and $Ca(H_2PO_4)_2/H_3PO_4$ solution were combined and stirred well; and the resulting mixture was taken to free-flowing dryness, by drying (ambient to about 200°C) overnight under a blanket of dry nitrogen with a slow nitrogen purge. The resulting material was thoroughly wetted with ethanol (a portion, sufficient to wet the solid, taken from a total of 150 mL); diethoxydiphenylsilane (1 g) was dissolved in the remainder of the total of 150 mL of ethanol to give a clear solution; the ethanol-wet $Ca(H_2PO_4)_2/H_3PO_4$-treated alumina and diethoxydiphenylsilane solution were combined and stirred well; and the resulting mixture was taken to free-flowing dryness, by drying (ambient to about 200°C) overnight under a blanket of dry nitrogen with a slow nitrogen purge.

Example 13

[203] **Catalyst 6** is made by adding 3.3 wt-% of Ca^{+2} onto alumina by incipient wetness impregnation. The impregnation solution is prepared by dissolving $Ca(HSO_4)_2$ and $Ca(SO_4)$ in water. The solution contains 0.066 g of Ca^{+2} in 1.3 mL solution. The clear solution is added to 2 g of alumina and mixed for about 5 min. The impregnated carrier is then calcined at 200 °C for 6 h to obtain the final catalyst.

Example 14

[204] This invention **Catalyst 7** is made by adding 2 wt-% of Ca^{+2} onto γ-alumina (powder, surface area >250 m^2/g) by incipient wetness impregnation. The carrier is first

calcined at 600 °C for 2 h. The impregnation solution is prepared by dissolving $Ca(SO_4)$ (136 mg) in water along with $NaHSO_4$ (142 mg). The solution contains 0.04 g of Ca^{+2} and an equal molar concentration of hydrogen sulfate and sulfate ions in 1.3 mL solution. The clear solution is added to 2 g of alumina and mixed for about 5 min. The impregnated carrier is then calcined at about 250 to 550 °C for 6 h to obtain the final catalyst.

Example 15

[205] This invention **Catalyst 8** is made by adding 2 wt-% of Ca^{+2} onto γ-alumina (powder, surface area >250 m^2/g) by incipient wetness impregnation. The carrier is first calcined at 600 °C for 2 h. The impregnation solution is prepared by dissolving $Ca(HPO_4)$, $Ca(H_2PO_4)_2$, and phosphoric acid (H_3PO_4) in a molar ratio of 5:5:1 in water. The solution contains 0.04 g of Ca^{+2} cations in 1.3 mL solution. The clear solution is added to 2 g of alumina and mixed for about 5 min. The impregnated carrier is then calcined at about 250 to 550 °C for 6 h to obtain the final catalyst.

Example 16

[206] This invention **Catalyst 9** is made by adding 2 wt-% of Ca^{+2} onto γ-alumina extrudate (1/16" diameter) by incipient wetness impregnation. The carrier is first calcined at 600 °C for 2 h. The impregnation solution is prepared by dissolving $Ca(HPO_4)$, $Ca(H_2PO_4)_2$, and phosphoric acid (H_3PO_4) in a molar ratio of 5:5:1 in water. The solution contains 0.4 g of Ca^{+2} cations in 13 mL solution. The clear solution is added to 20 g of alumina and mixed for about 5 min. The impregnated carrier is then calcined at about 250 to 550 °C for 6 h to obtain the final catalyst.

Example 17

[207] This invention **Catalyst 10** is made by adding 2 wt-% of Ca^{-2} onto γ-alumina extrudate (1/8" diameter) by incipient wetness impregnation. The carrier is first calcined at 600 °C for 2 h. The impregnation solution is prepared by dissolving $Ca(SO_4)$ (1.36 g) in water along with $NaHSO_4$ (1.42 g). The solution contains 0.4 g of Ca^{+2} and an equal molar concentration of hydrogen sulfate and sulfate ions in 13 mL solution. The clear solution is added to 20 g of alumina extrudate and mixed for about 5 min. The impregnated carrier is then calcined at about 250 to 550 °C for 6 h to obtain the final catalyst.

Example 18

[208] This invention **Catalyst 11** is made by adding 2 wt-% of Ca^{-2} onto γ-alumina extrudate (1/8" diameter) by incipient wetness impregnation. The carrier is first calcined at 600 °C for 2 hr. The impregnation solution is prepared by dissolving $Ca(SO_4)$ (1.36 g) in water along with $NaHSO_4$ (1.42 g). The solution contains 0.4 g of Ca^{+2} and an equal molar concentration of hydrogen sulfate and sulfate ions in 13 mL solution. The clear solution is added to 20 g of alumina extrudate and mixed for about 5 min. The impregnated carrier is then calcined at about 250 to 550 °C for 6 h to obtain the final catalyst. This material is treated with an ethanol (15 mL) solution containing $Ph_2Si(OEt)_2$ (0.74 g) and mixed for 5 min and then placed under reduced pressure (400 torr) and heated to 50 °C for 2 hr and then heated to 200 °C for 4 hr.

Example 19

Conversion of crotonaldehyde to crotyl alcohol in Clostridium fermentation

Microorganism and culture medium

[209] ABE-producing *Clostridium saccharobutylicum* strains were used for the following experiments, including publicly available *Clostridium saccharobutylicum* P262, to demonstrate the fermentability of crotyl alcohol.

[210] J2 Media with 6% glucose was used to grow *Clostridium* cells for crotyl alcohol production. Pre-seed was inoculated with 0.4% (v/v) of cells from glycerol stock. The inoculum transfer from pre-seed to seed tank was made to achieve a final OD of 1 (equal to 0.32 g of dry cell weight per L of broth) in the seed tanks.

[211] For confirming the *adh* activity, the microbial cultures were spiked with different concentrations of crotonaldehyde during the solventogenesis stage. Spiking concentrations of crotonaldehyde ranged from 0 to 2 g/L. Since crotonaldehyde is very toxic to the microorganism, fed-batch fermentation was also done to test the tolerance of the microorganism. Small amount of crotonaldehyde were slowly added into fermentation to final concentration of 2 g/L over 6 hours. Samples were taken immediately and 24 hours after crotonaldehyde addition.

Alcohol Dehydrogenase Activity

[212] Fig. 24 shows the production of crotyl alcohol in the microbial cultures. The microorganism was able to take up crotonaldehyde and produce crotyl alcohol. The

reaction is relatively fast because crotonaldehyde is toxic to the microorganism, which is shown in Fig. 25 with no more butanol produced after the addition of crotonaldehyde at the concentration higher than 1 g/L.

[213] Fig. 26 shows crotyl alcohol production with different concentrations of crotonaldehyde addition.

[214] Where a range of values is provided, it is understood that each intervening value, to the tenth of the unit of the lower limit unless the context clearly dictates otherwise, between the upper and lower limits of that range is also specifically disclosed. Each smaller range between any stated value or intervening value in a stated range and any other stated or intervening value in that stated range is encompassed within the invention. The upper and lower limits of these smaller ranges may independently be included or excluded in the range, and each range where either, neither or both limits are included in the smaller ranges is also encompassed within the invention, subject to any specifically excluded limit in the stated range. Where the stated range includes one or both of the limits, ranges excluding either or both of those included limits are also included in the invention.

[215] While the invention has been described, disclosed, illustrated and shown in various terms of certain embodiments or modifications which it has presumed in practice, the scope of the invention is not intended to be, nor should it be deemed to be, limited thereby and such other modifications or embodiments as may be suggested by the teachings herein are particularly reserved especially as they fall within the breadth and scope of the claims here appended.

[216] Although the foregoing invention has been described in some detail by way of illustration and examples for purposes of clarity of understanding, it will be apparent to those skilled in the art that certain changes and modifications may be practiced without departing from the spirit and scope of the invention. Therefore, the description should not be construed as limiting the scope of the invention.

[217] All publications, patents, and patent applications cited herein are hereby incorporated by reference in their entireties for all purposes and to the same extent as if each individual publication, patent, or patent application were specifically and individually indicated to be so incorporated by reference.

CLAIMS

We claim:

1. A method for dehydrating an alkenol or a diol to a diene, comprising:

contacting and heating said alkenol or diol with at least one catalyst that comprises an inorganic support, one or more promoter(s) selected from one or more Group 1 element(s), one or more Group 2 element(s), and combinations thereof, and one or more buffer-adjusted anion(s) at a WHSV at least about 0.3 hr^{-1},

thereby producing a diene product that comprises one or more diene(s) at a one-pass yield greater than about 73 mol%.

2. A method according to claim 1, wherein said WHSV is about 0.3 hr^{-1} to about 10 hr^{-1}.

3. A method according to claim 1, wherein said alkenol or diol is in a mixture with about 1 ppm to about 93 wt% water.

4. A method according to claim 1, wherein the selectivity of diene production is at least about 75%.

5. A method according to claim 1, wherein said alkenol is a bio-alkenol.

6. A method according to claim 1, wherein said alkenol is crotyl alcohol.

7. A method according to claim 1, wherein said alkenol is bio-crotyl alcohol.

8. A method according to claim 1, wherein said alkenol is a C_4 to C_6 alkenol

9. A method according to claim 1, wherein said one or more diene(s) is 1,3-butadiene.

10. A method according to claim 4, wherein the selectivity of diene production is greater than about 95%.

11. A method according to claim 4, wherein the selectivity of diene production is about 90% to about 99%.

12. A method according to claim 1, wherein one-pass conversion of said alkenol or diol is greater than about 95 mol%.

13. The method according to claim 3, wherein said mixture comprises about 15 to about 70 wt% water.

14. A method according to claim 1, wherein said inorganic support comprises γ-

alumina.

15. A method according to claim 1, wherein said catalyst is further modified by treatment with at least one organosilane.

16. A method according to claim 15, wherein said organosilane comprises an alkoxysilane.

17. A method according to claim 15, wherein said organosilane comprises a chlorosilane.

18. A method according to claim 1, further comprising providing at least one carrier gas.

19. A method according to claim 18, wherein said carrier gas comprises nitrogen and/or argon, and mixtures thereof.

20. A method according to claim 1, wherein said heating comprises a temperature of about 200° C to about 440° C.

21. The method according to claim 20, wherein said temperature comprises about 250° C to about 320° C.

22. A method according to claim 1, further comprising separating water from said diene product, thereby producing a dry diene product.

23. A method according to claim 9, comprising contacting said 1,3-butadiene product with a second catalyst under conditions sufficient to form 1-butene.

24. A method according to claim 23, wherein said 1-butene is contacted with a third catalyst selected from a Ziegler-Natta catalyst, a mesoporous oligomerization catalyst, and an acid catalyst under conditions sufficient to form oligomers.

25. A method according to claim 24, further comprising using said oligomers to produce a diesel fuel.

26. A method according to claim 25 wherein said diesel fuel comprises a flashpoint of about 38° C to about 100° C, a Cetane rating of about 45 to about 60, and aromatic content of less than about 0.5 wt-%.

27. A method according to claim 24, further comprising using said oligomers to produce a jet fuel.

28. A method according to claim 27, wherein said jet fuel comprises a flashpoint of

about 38° C to about 100° C, a cold flow viscosity of less than about 8.0 cSt at -20 °C, and aromatic content of less than about 0.5 wt-%.

29. A method according to claim 1, wherein the promoter is selected from Mg, Ca, Sr, Ba, and combinations thereof.

30. A method according to claim 29, wherein the promoter is Ca.

31. A method according to claim 1, wherein the buffer-adjusted anion is selected from SO_4^{-2}, $H_2PO_4^{-1}$, and mixtures thereof.

32. A method for dehydrating an alkenol or a diol to a diene, comprising:

contacting and heating said alkenol or diol with at least one catalyst that comprises a calcium-modified inorganic support and one or more buffer-adjusted anion(s),

thereby producing a diene product that comprises one or more diene(s) at a one-pass yield greater than about 80 mol%.

33. A method according to claim 32, wherein said alkenol is a bio-alkenol.

34. A method according to claim 32, wherein said alkenol is crotyl alcohol.

35. A method according to claim 32, wherein said alkenol is bio-crotyl alcohol.

36. A method according to claim 32, wherein the alkenol is a C_4 to C_6 alkenol

37. A method according to claim 32, wherein said diene product is 1,3-butadiene.

38. A method according to claim 32, wherein said alkenol is in a mixture with about 1 ppm to about 93 wt-% water.

39. A method according to claim 32, wherein the contacting is at a WHSV of about 0.3 hr^{-1} to about 10 hr^{-1}.

40. A method according to claim 32, wherein said catalyst is further modified by treatment with at least one organosilane.

41. A catalyst composition for producing dienes from alkenols or diols with high selectivity, comprising:

an inorganic support that is modified with at least one Group 1 element, at least one Group 2 element, or a combination thereof;

one or more buffer-adjusted anion;

and optionally, at least one organosilane.

42. A catalyst composition according to claim 41, comprising at least one Group 2 element selected from Mg, Ca, Sr, and Ba.

43. A catalyst composition according to claim 42, wherein the Group 2 element is Ca.

44. A catalyst composition according to claim 41, wherein the buffer-adjusted anion is selected from SO_4^{-2}, $H_2PO_4^{-1}$, and mixtures thereof.

45. A catalyst composition according to claim 41, wherein the buffer-adjusted anion is $H_2PO_4^{-1}$, wherein $H_2PO_4^{-1}$ is a component of a mixture that further optionally comprises one or more additional anions selected from HPO_4^{-2} and PO_4^{-3} and one or more charge balancing counter cations selected from H^{+1}, Li^{+1}, Na^{+1}, K^{+1}, Mg^{+2}, Ca^{+2}, Sr^{+2}, Ba^{+2}, and combinations thereof.

46. A catalyst composition according to claim 41, comprising an organosilane, wherein the organosilane is diethoxydiphenylsilane.

47. A catalyst composition according to claim 41, wherein the inorganic support is γ-alumina or zinc aluminate.

48. A method of making a catalyst composition, comprising:

a) modifying an inorganic support with at least one Group 2 element and at least one buffer-adjusted anion to produce a first modified inorganic support, wherein the first modified inorganic support comprises said at least one Group 2 element and said at least one buffer-adjusted anion;

b) calcining the first modified inorganic support to produce a calcined first modified inorganic support;

c) contacting the calcined first modified inorganic support with at least one organosilane to produce a second modified inorganic support, wherein the second modified inorganic support comprises said at least one Group 2 element, said at least one buffer-adjusted anion, and said at least one organosilane, or fragment thereof; and

d) calcining the second modified inorganic support to produce the catalyst composition.

49. A method for dehydrating crotyl alcohol to a diene product, comprising:

contacting and heating said crotyl alcohol with at least one catalyst that

comprises an inorganic support, one or more promoter(s) selected from one or more Group 1 element(s), one or more Group 2 element(s), and combinations thereof, and one or more buffer-adjusted anion(s) at a WHSV at least about 0.3 hr^{-1},

thereby producing a diene product that comprises one or more diene(s) at a one-pass yield greater than about 73 mol%.

50. A method according to claim 49, wherein said crotyl alcohol is in a mixture comprising about 1 ppm to about 93 wt-% water.

51. The method according to claim 50, wherein said mixture comprises about 5 wt-% to about 70 wt-% water.

52. A method according to claim 49, wherein said crotyl alcohol is bio-crotyl alcohol.

53. A method according to claim 49, wherein said one or more diene(s) is 1,3-butadiene.

54. A method according to claim 49, wherein the molar selectivity of said one or more diene(s) production is greater than about 95%.

55. A method according to claim 49, wherein the molar selectivity of said one or more diene(s) production is about 90% to about 99%.

56. A method according to claim 49, wherein said inorganic support comprises γ-alumina.

57. A method according to claim 49, wherein said catalyst is further modified by treatment with at least one organosilane.

58. The method according to claim 57, wherein said organosilane comprises an alkoxysilane.

59. A method according to claim 49, further comprising providing at least one carrier gas.

60. A method according to claim 49, wherein the Group 2 element is Ca.

61. A method according to claim 49, wherein the buffer-adjusted anion is selected from SO_4^{-2}, $H_2PO_4^{-1}$, and mixtures thereof.

62. A method of producing crotyl alcohol, comprising growing a microorganism in a growth medium under fermentation conditions suitable for production of crotyl alcohol.

63. A method according to claim 62, wherein the microorganism expresses a native

enzyme that can produce crotyl alcohol from a substrate in the growth medium.

64. A method according to claim 63, wherein the enzyme is an alcohol and/or aldehyde dehydrogenase.

65. A method according to claim 63, wherein the enzyme is an acyl-CoA reductase.

66. A method according to claim 63, wherein the substrate is crotonaldehyde.

67. A method according to claim 63, wherein the crotyl alcohol is produced from upstream biosynthetic intermediates comprising crotonic acid, crotonyl-CoA and/or other intermediates in the butanol/butanoate pathway.

68. A method according to claim 63, wherein the native enzyme is part of a native butanoate/butanol, fatty acid, amino acid degradation, or related metabolic pathway in the microorganism.

69. A method according to claim 68, wherein the metabolic pathway comprises an enzyme of a glycolytic or pentose phosphate pathway that converts glucose and/or xylose to acetyl-CoA, a thiolase enzyme that converts acetyl-CoA to acaetoacetyl-CoA, a 3-hydroxy butyryl CoA dehydrogenase enzyme that converts acetoacetyl-CoA to 3-hydroxybutyryl-CoA, a crotonase enzyme that converts 3-hydroxybutyryl-CoA to crotonyl-CoA, an aldehyde dehydrogenase enzyme that converts crotonyl-CoA to crotonaldehyde, and an alcohol dehydrogenase enzyme that converts crotonaldehyde to crotyl alcohol._

70. A method according to claim 62, wherein the microorganism is a *Clostridium* strain.

71. A method according to claim 70, wherein the *Clostridium* microorganism expresses an enzyme that converts crotonaldehyde to crotyl alcohol.

72. A method according to claim 70, wherein the *Clostridium* microorganism expresses an enzyme that converts crotonyl-CoA to crotonaldehyde or crotyl alcohol.

73. A method according to claim 71, wherein the enzyme that converts crotonaldehyde to crotyl alcohol is a native enzyme in the *Clostridium* microorganism.

74. A method according to claim 72, wherein the enzyme that converts crotonyl-CoA to crotonaldehyde or crotyl alcohol is a native enzyme in the *Clostridium* microorganism.

75. A method according to claim 71, wherein the enzyme is an alcohol and/or aldehyde dehydrogenase.

76. A method according to claim 72, wherein the enzyme is an acyl-CoA reductase that

converts crotonyl-CoA to crotyl alcohol without intervening additional enzymatic activities

77. A method according to claim 62, wherein the growth medium comprises one or more substances that increase production of crotyl alcohol in comparison to a growth medium that does not comprise the one or more substances.

78. A method according to claim 77, wherein the one or more substances comprise one or more co-factors of enzymes in a butanoate/butanol pathway in the microorganism.

79. A method according to claim 78, wherein the one or more co-factors include one or more of Mn, Fe, NADPH, and NADP.

80. A method according to claim 78, wherein the one or more substances modifies the activity of one or more native enzyme(s) in the microorganism, thereby increasing crotyl alcohol production.

81. A method according to claim 80, wherein bcd is inhibited by the one or more substances in the growth medium.

82. A method according to claim 80, wherein adhE and adh2 are activated by the one or more substances in the growth medium.

83. A method according to claim 77, wherein the one or more substances in the growth medium comprises one or more divalent metal ion(s), iodoacetamide, myristoyl-CoA, N-ethylmaleimide, and/or palmitoyl-CoA.

84. A method according to claim 83, wherein the one or more divalent metal ion(s) is selected from selected from Ca, Co, Zn, and Mn, and combinations thereof.

85. A method according to claim 62, wherein the growth medium comprises carbohydrate molecules produced by hydrolysis of lignocellulosic biomass.

86. An engineered *Clostridium* strain that produces a greater amount of crotyl alcohol than the parent strain from which it was derived.

87. A *Clostridium* strain according to claim 86, wherein said strain has been engineered to comprise a reduced level or activity of butyryl-CoA dehydrogenase in comparison to the parent strain from which it was derived.

88. A *Clostridium* strain according to claim 87, wherein one or more enzyme(s) of a crotyl alcohol biosynthetic pathway is overexpressed.

89. A *Clostridium* strain according to claim 88, wherein the one or more enzyme(s) of

crotyl alcohol biosynthesis comprises an enzyme of a glycolytic or pentose phosphate pathway that converts glucose and/or xylose to acetyl-CoA, a thiolase enzyme that converts acetyl-CoA to acaetoacetyl-CoA, a 3-hydroxy butyryl CoA dehydrogenase enzyme that converts acetoacetyl-CoA to 3-hydroxybutyryl-CoA, a crotonase enzyme that converts 3-hydroxybutyryl-CoA to crotonyl-CoA, an aldehyde dehydrogenase enzyme that converts crotonyl-CoA to crotonaldehyde, and/or an alcohol dehydrogenase enzyme that converts crotonaldehyde to crotyl alcohol.

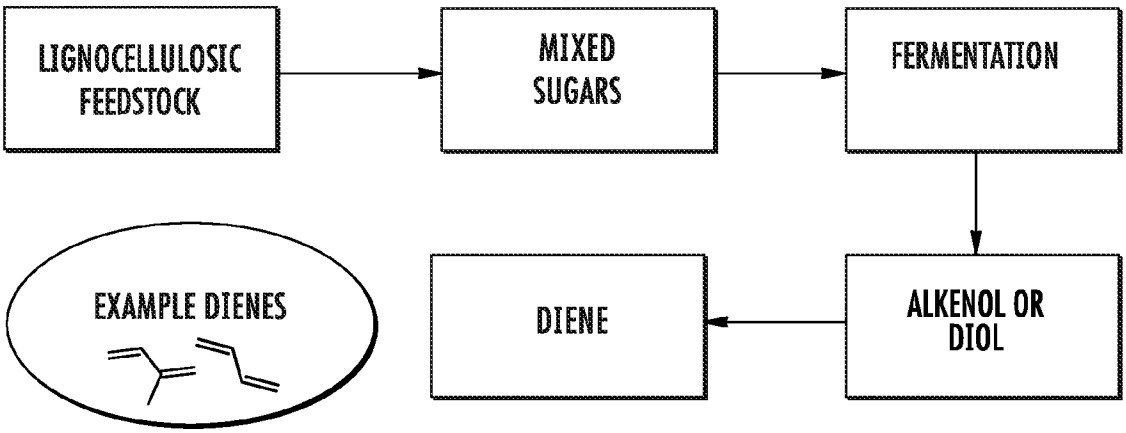
90. A *Clostridium* strain according to claim 89, wherein the one or more enzyme(s) of crotyl alcohol biosynthesis comprises at least one alcohol dehydrogenase.

91. A *Clostridium* strain according to claim 87, wherein said strain is an SiHybrid in which butyryl-CoA dehydrogenase activity is downregulated in comparison to the parent strain from which it was derived.

92. A *Clostridium* strain according to claim 87, wherein butyryl-CoA dehydrogenase has been inactivated using a targetron gene knockout system.

93. A method of producing crotyl alcohol, comprising growing the *Clostridium* strain according to claim 86 in a growth medium under fermentation conditions suitable for production of crotyl alcohol.

94. A method according to claim 93, wherein the growth medium comprises carbohydrate molecules produced by hydrolysis of lignocellulosic biomass.



NON-FOOD BIOMASS CONVERSION TO DIENES

FIG. 1

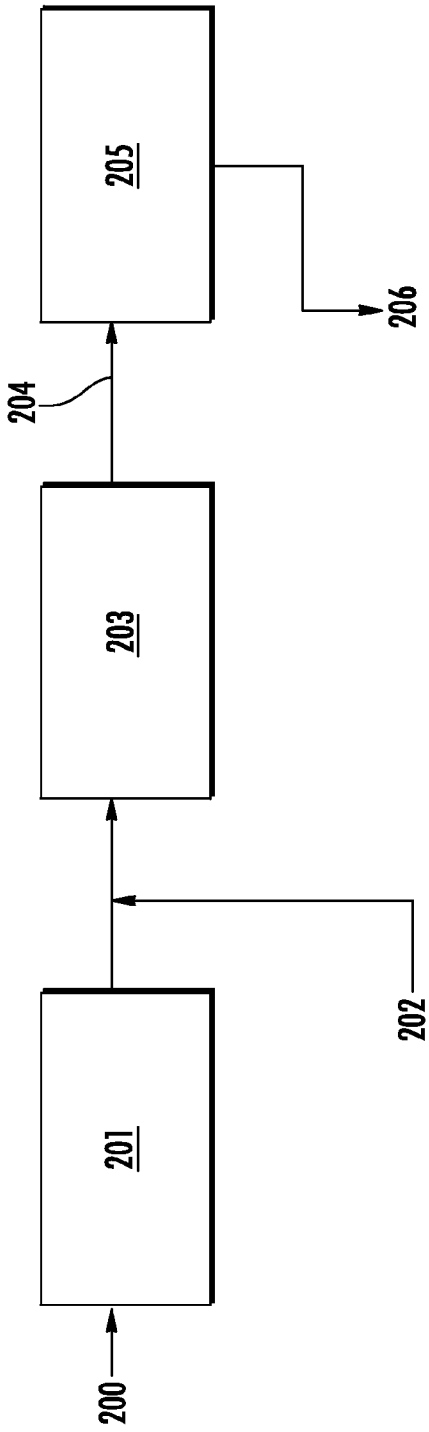


FIG. 2

9/13/2020, EAST Version: 4.1.7.1

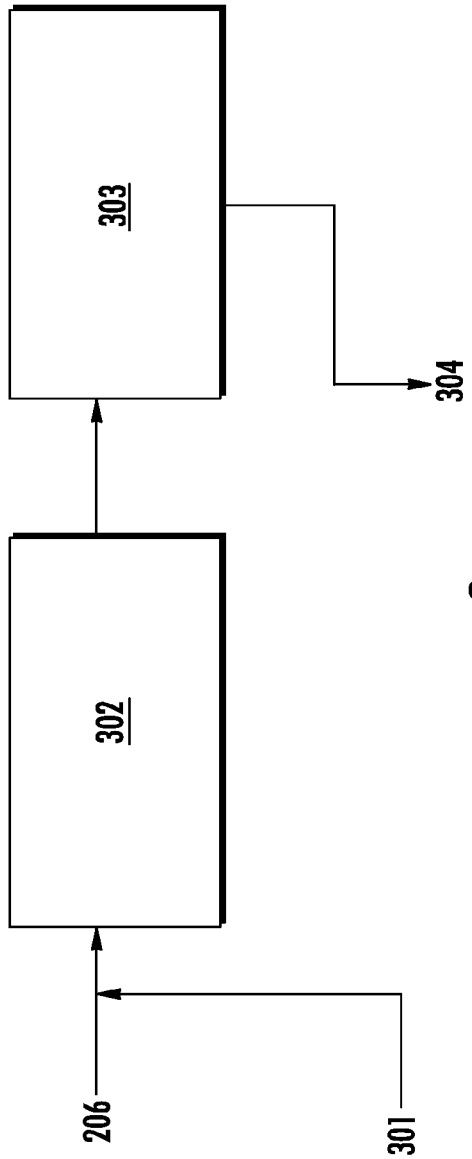


FIG. 3

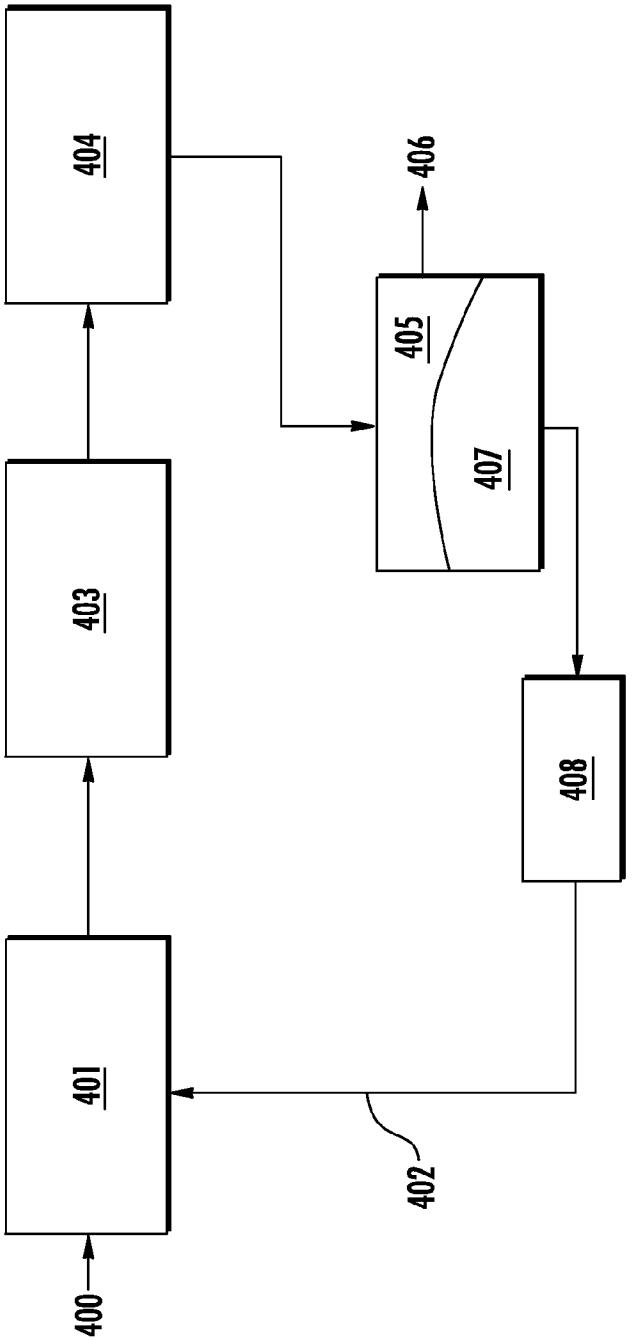


FIG. 4

9/13/2020, EAST Version: 4.1.7.1

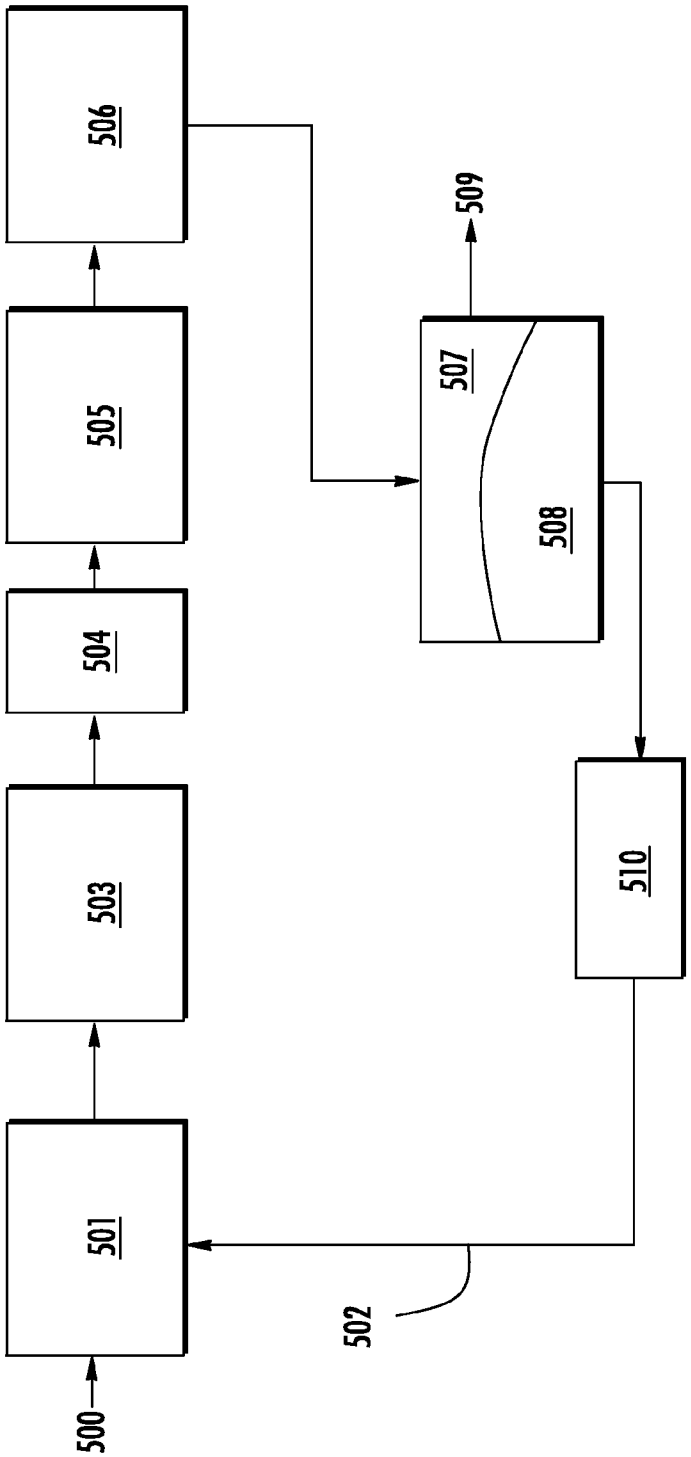


FIG. 5

9/13/2020, EAST Version: 4.1.7.1

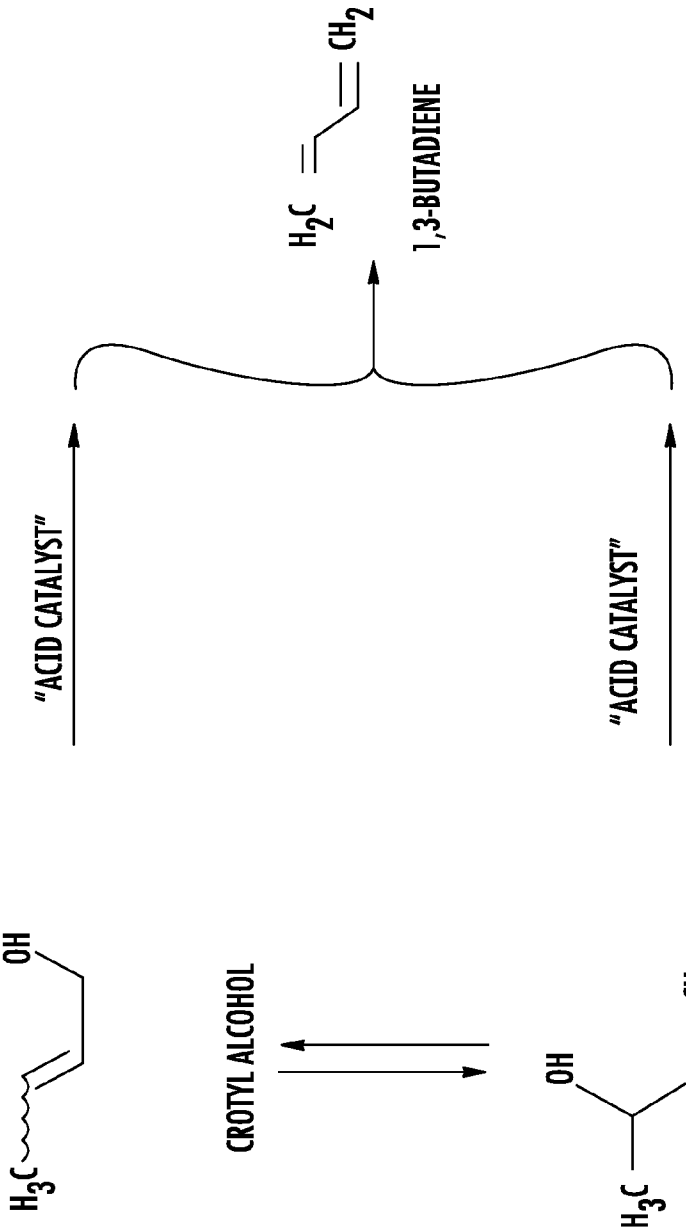


FIG. 6

9/13/2020, EAST Version: 4.1.7.1

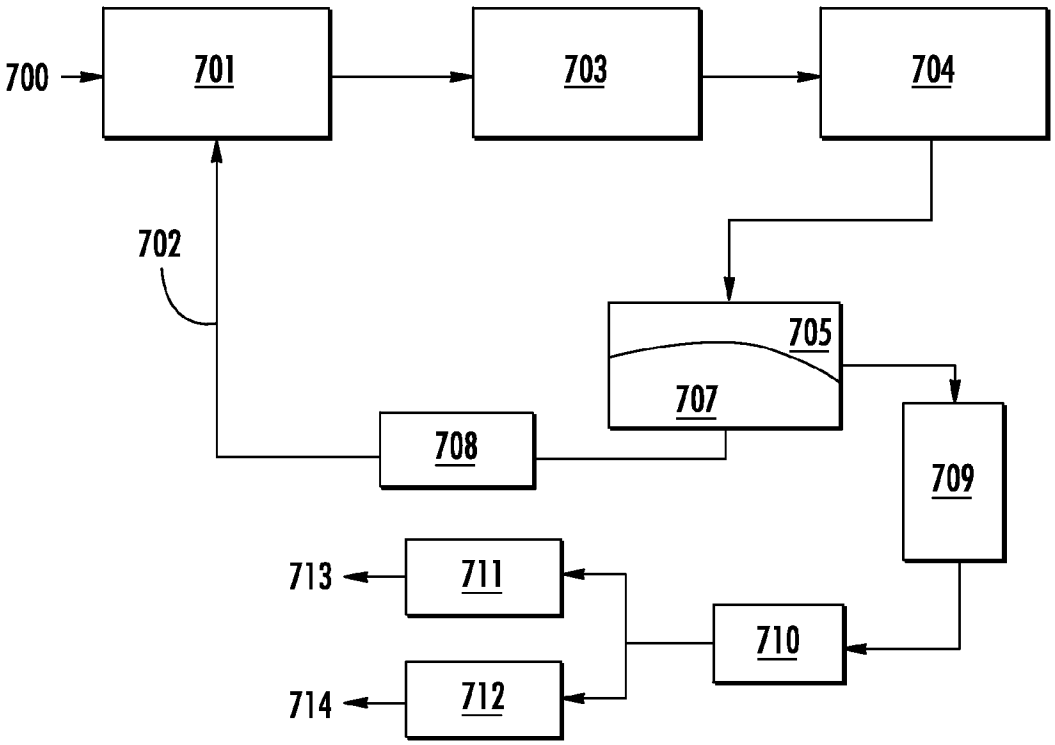


FIG. 7

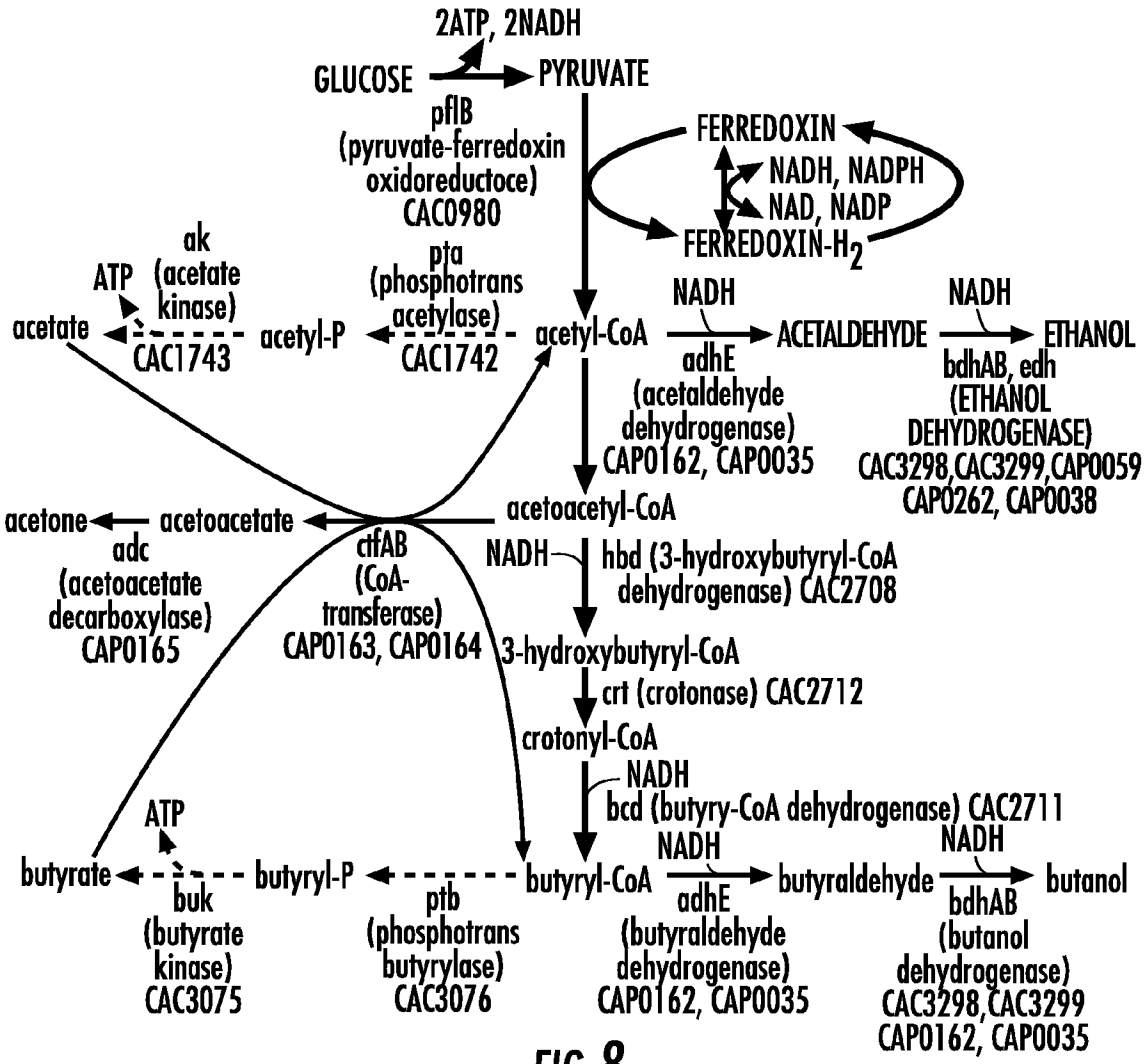
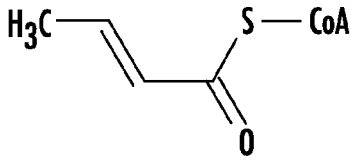


FIG. 8



CROTONYL-CoA

FIG. 9

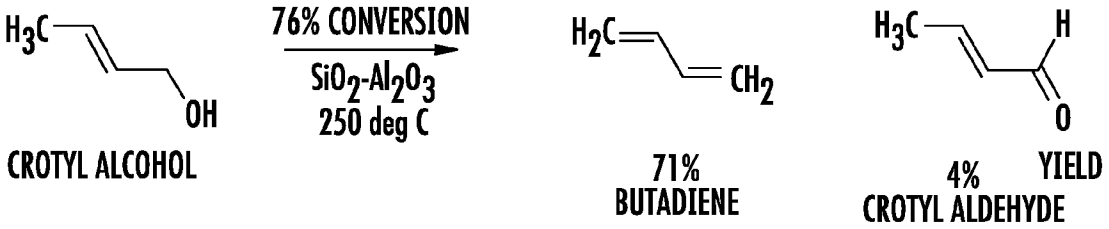

H3C
OH
CROTYL ALCOHOL
76% CONVERSION
SiO2-Al2O3
250 deg C
H2C
CH2
71%
BUTADIENE
H3C
H
O
4%
CROTYL ALDEHYDE
YIELD

FIG. 10

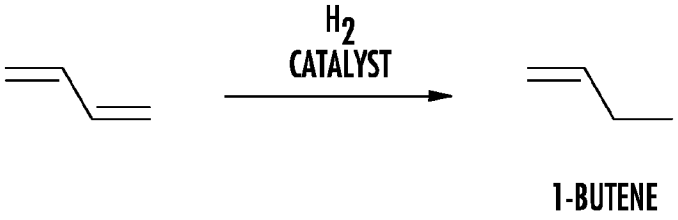

H2
CATALYST
1-BUTENE

FIG. 11

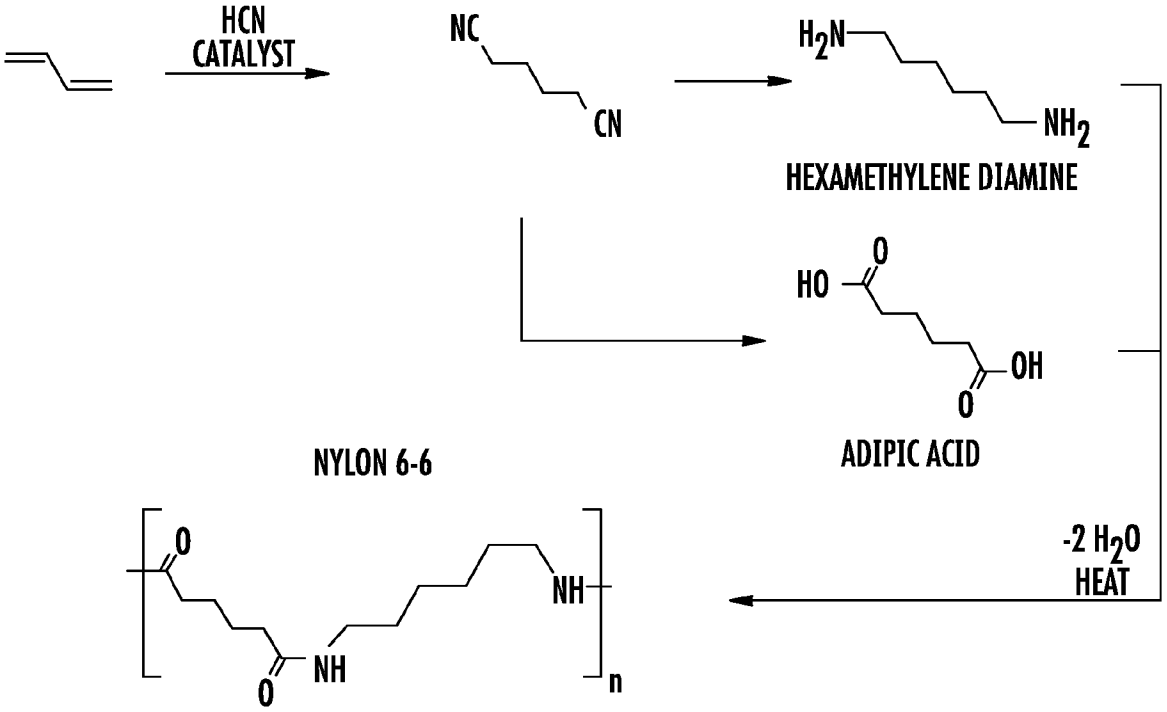


FIG. 12

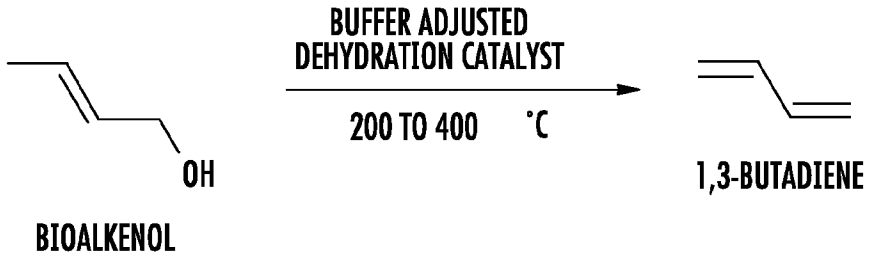


FIG. 13

9/13/2020, EAST Version: 4.1.7.1

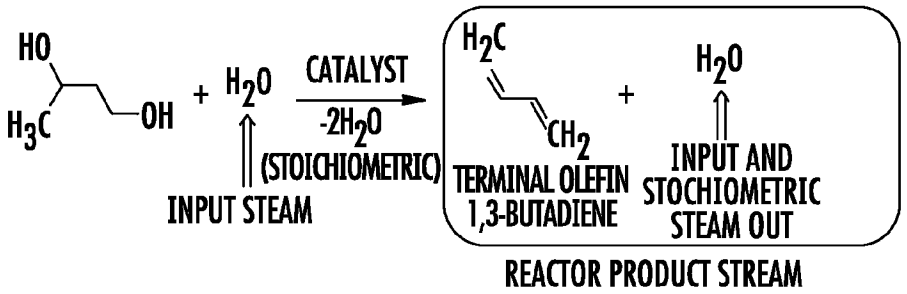


FIG. 14

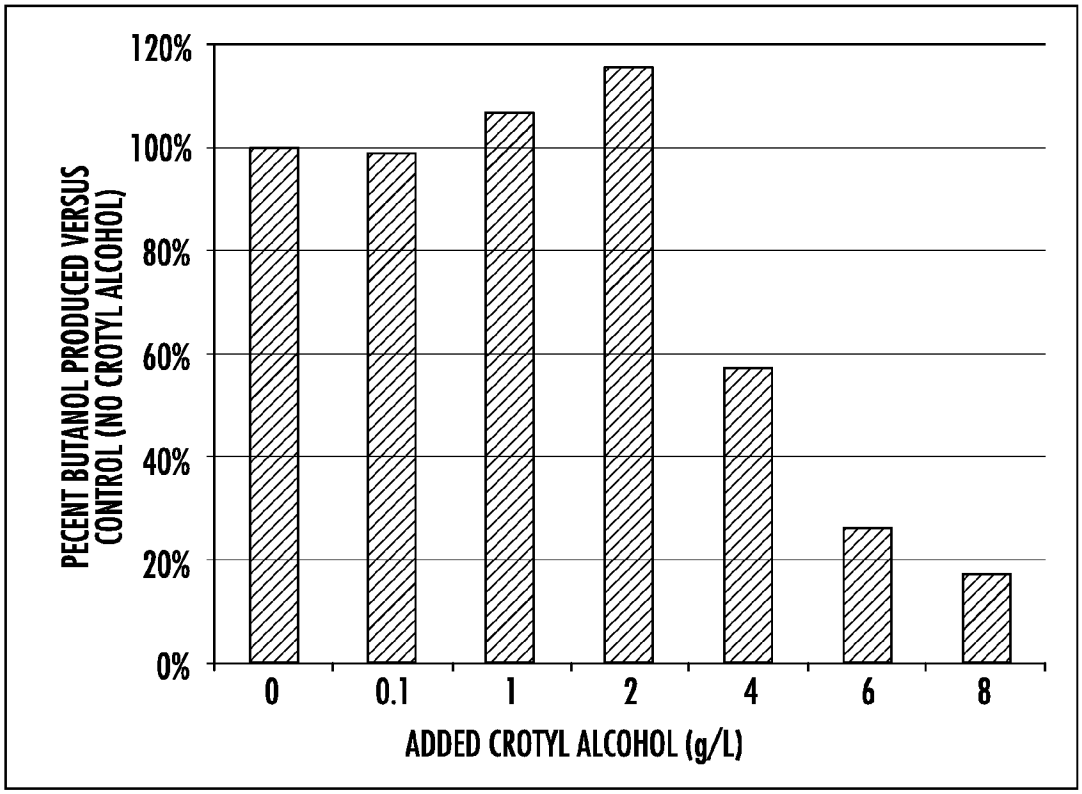


FIG. 15

9/13/2020, EAST Version: 4.1.7.1

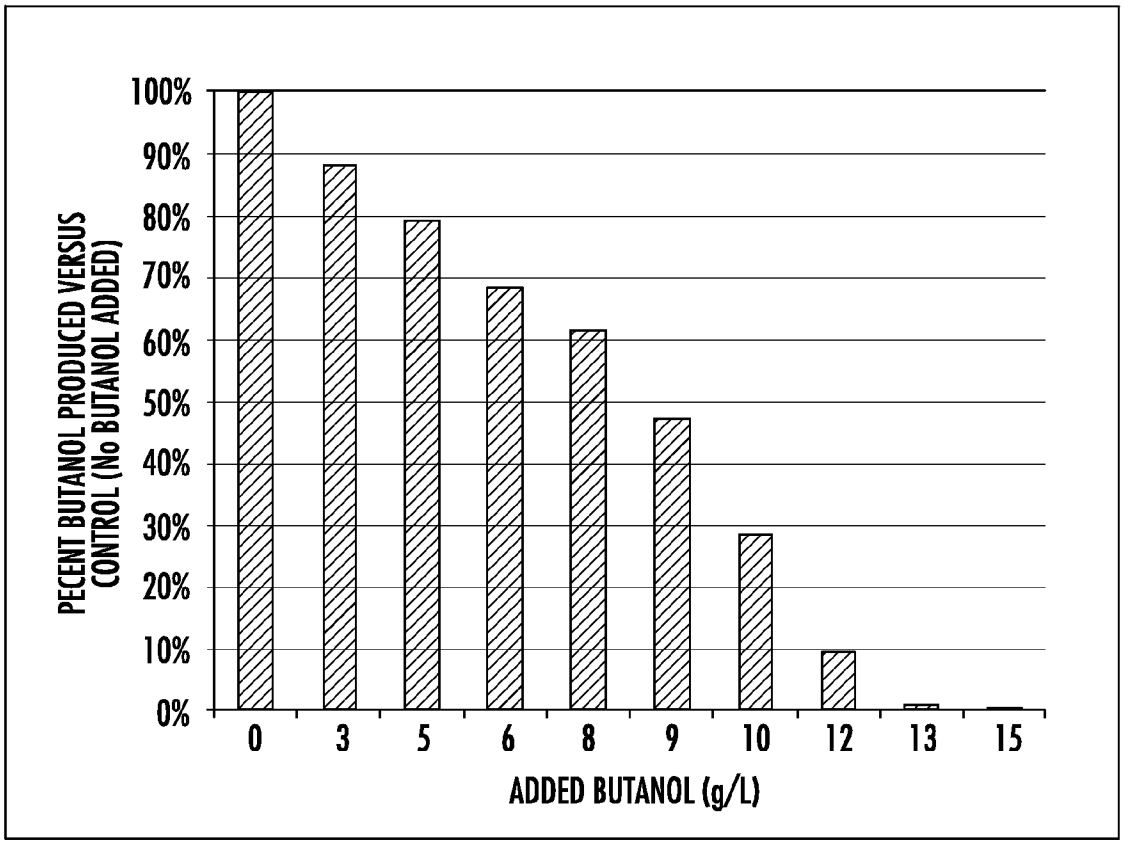


FIG. 16

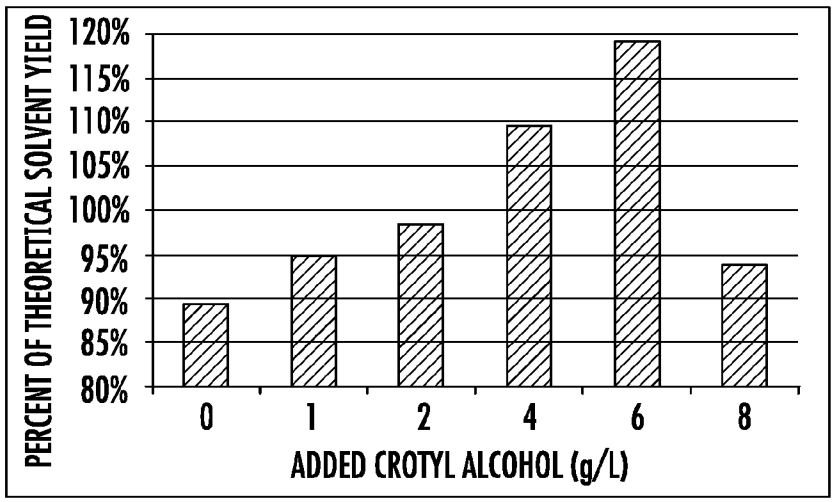


FIG. 17

9/13/2020, EAST Version: 4.1.7.1

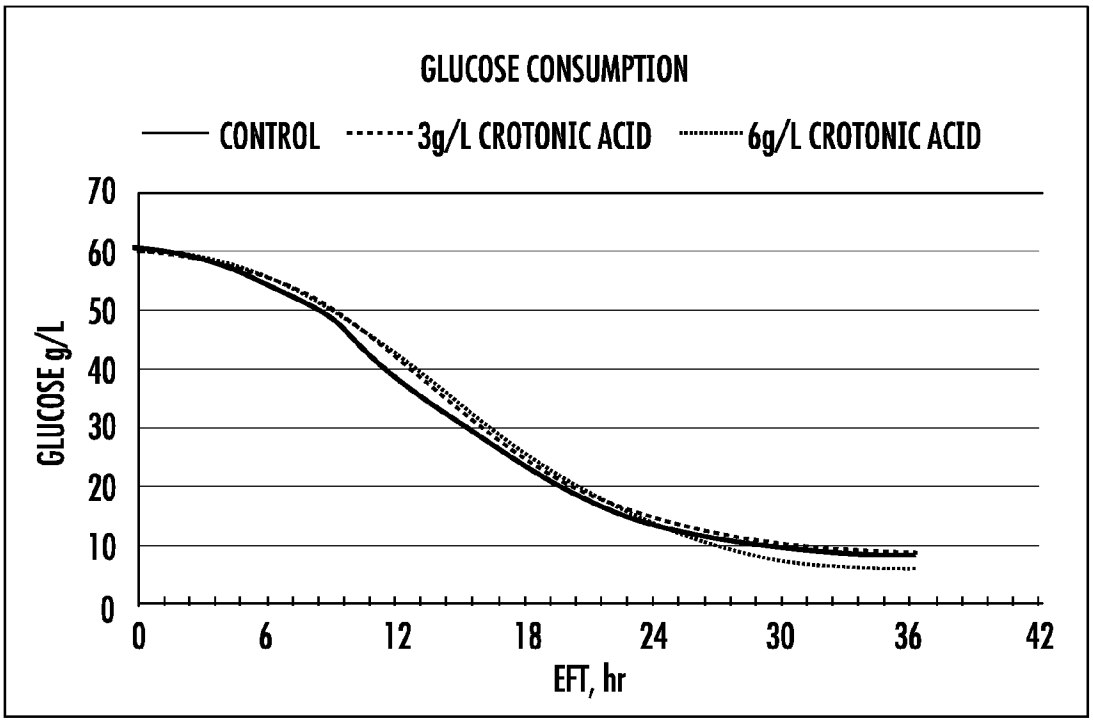


FIG. 18

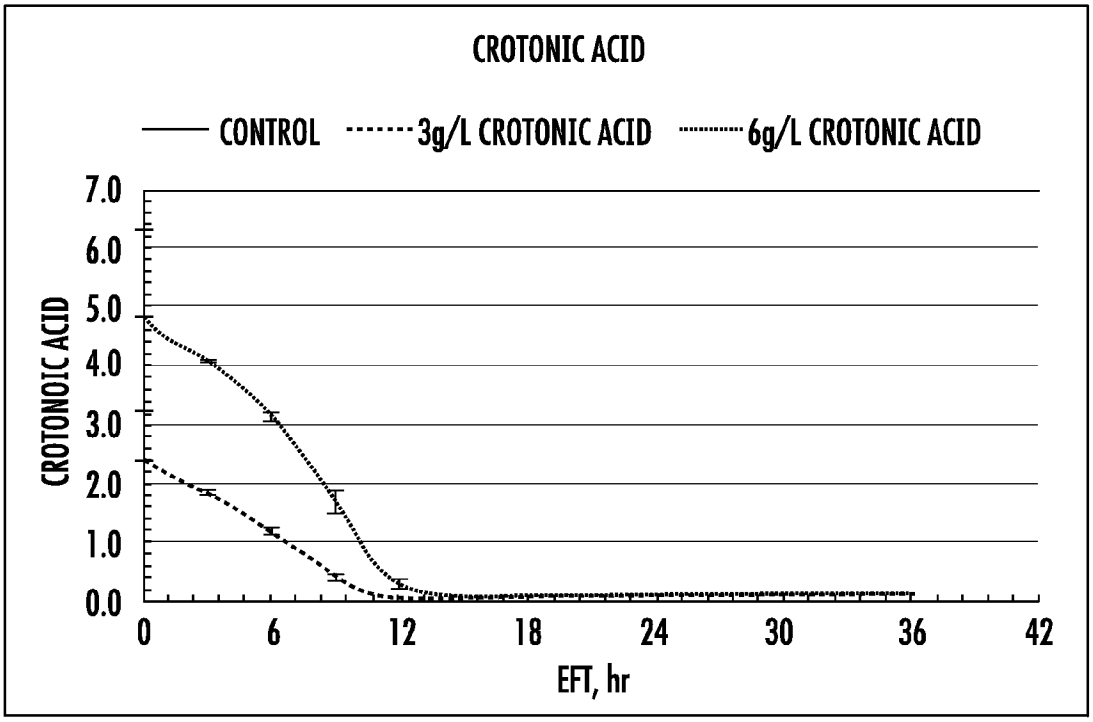


FIG. 19

9/13/2020, EAST Version: 4.1.7.1

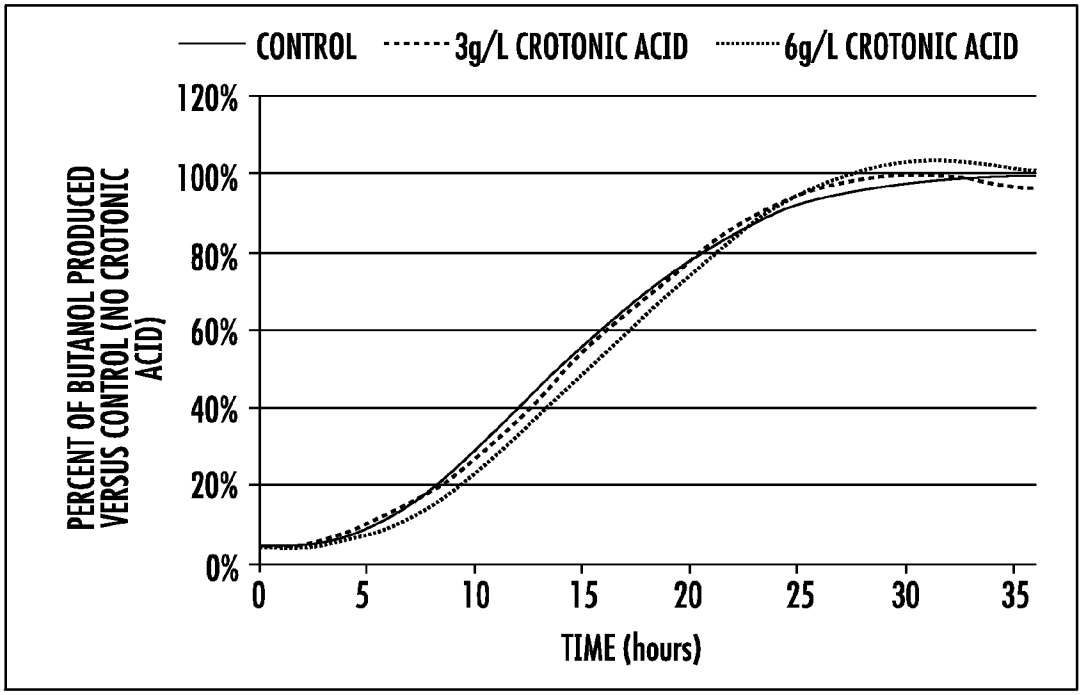

CONTROL
3g/L CROTONIC ACID
6g/L CROTONIC ACID
120%
100%
80%
60%
40%
20%
0%
PERCENT OF BUTANOL PRODUCED VERSUS CONTROL (NO CROTONIC ACID)
0
5
10
15
20
25
30
35
TIME (hours)

FIG. 20

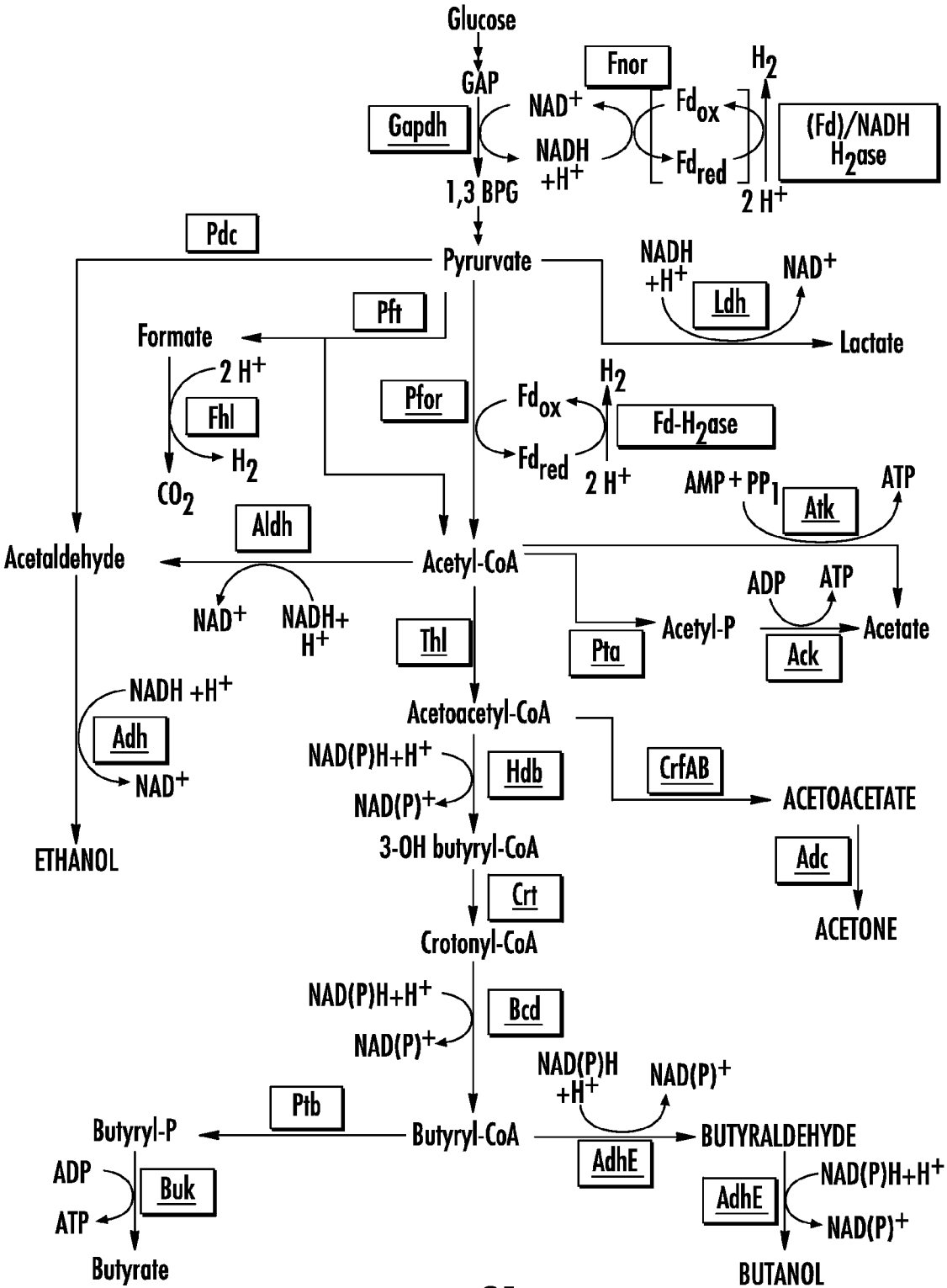


FIG. 21

9/13/2020, EAST Version: 4.1.7.1

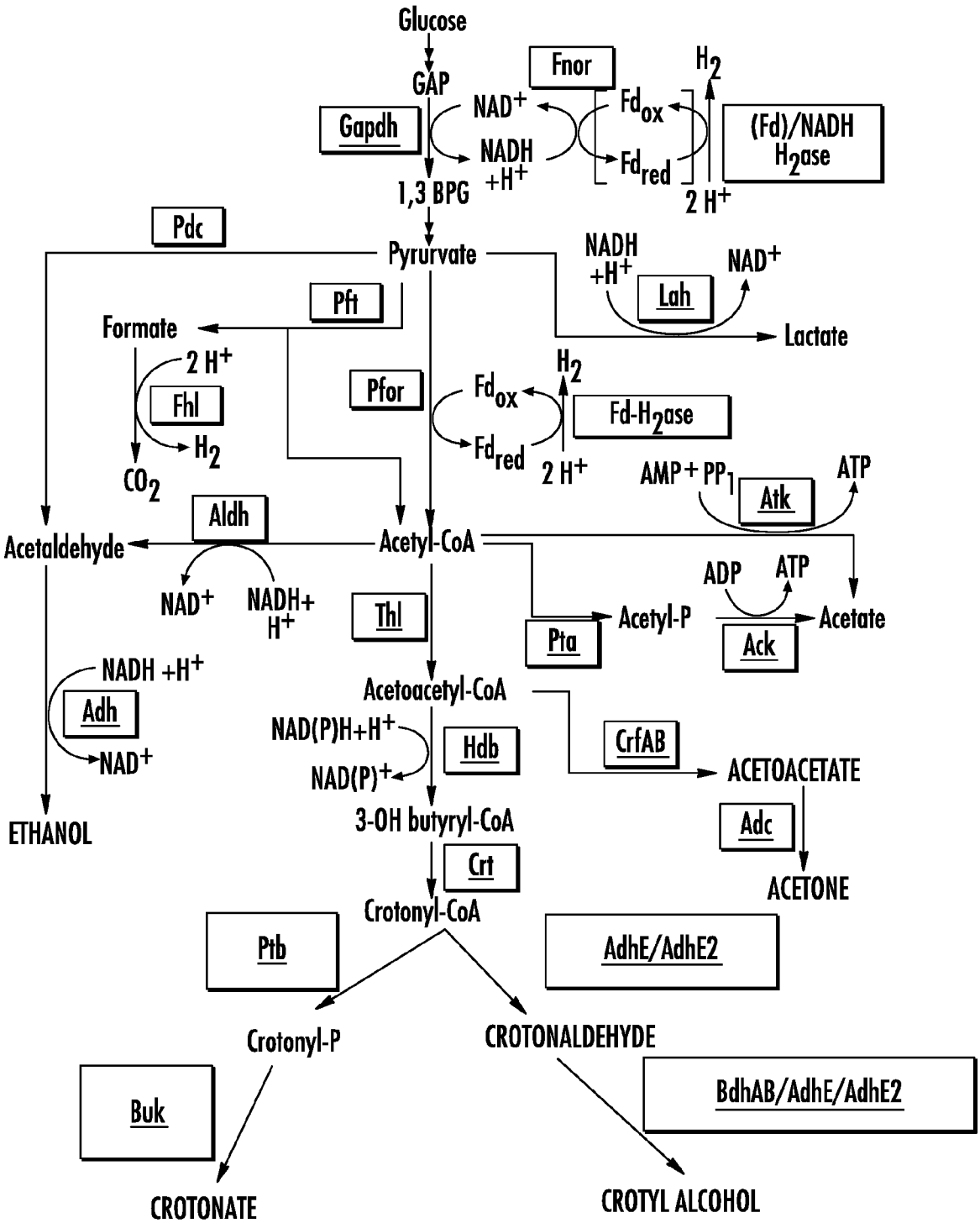

Glucose
GAP
Gapdh
NAD+
NADH +H+
1,3 BPG
Fnor
Fdox
Fdred
H2
2 H+
(Fd)/NADH H2ase
Pdc
Pyrurvate
NADH +H+
NAD+
Lah
Lactate
Pfl
Formate
2 H+
Fhl
H2
CO2
Pfor
Fdox
Fdred
H2
2 H+
Fd-H2ase
AMP + PP1
Atk
ATP
Aldh
Acetaldehyde
Acetyl-CoA
NAD+
NADH+ H+
Thl
ADP
ATP
Pta
Acetyl-P
Ack
Acetate
NADH +H+
Adh
NAD+
ETHANOL
Acetoacetyl-CoA
NAD(P)H+H+
NAD(P)+
Hdb
CrfAB
ACETOACETATE
Adc
ACETONE
3-OH butyryl-CoA
Crt
Crotonyl-CoA
Ptb
AdhE/AdhE2
Crotonyl-P
CROTONALDEHYDE
Buk
BdhAB/AdhE/AdhE2
CROTONATE
CROTYL ALCOHOL

FIG. 22

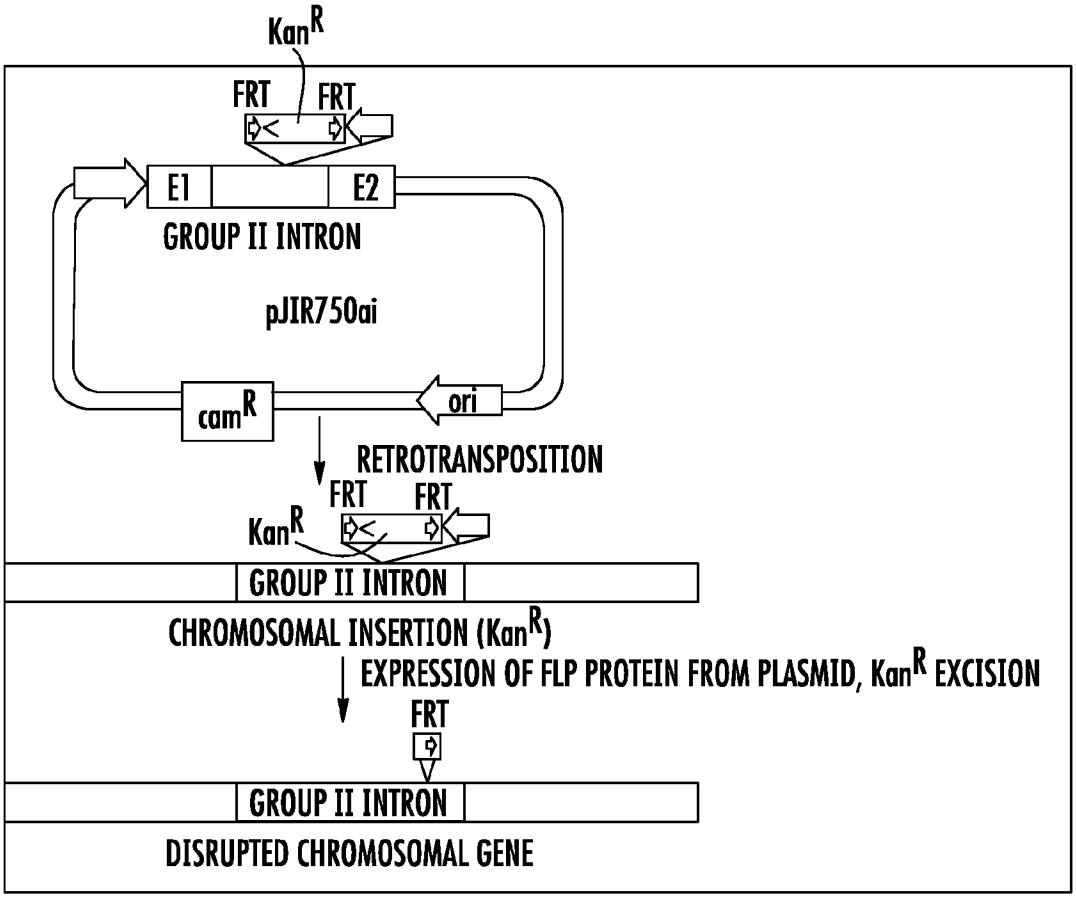

Kan^R
FRT
FRT
E1
E2
GROUP II INTRON
pJIR750ai
cam^R
ori
RETROTRANSPOSITION
FRT
FRT
Kan^R
GROUP II INTRON
CHROMOSOMAL INSERTION (Kan^R)
EXPRESSION OF FLP PROTEIN FROM PLASMID, Kan^R EXCISION
FRT
GROUP II INTRON
DISRUPTED CHROMOSOMAL GENE

FIG. 23

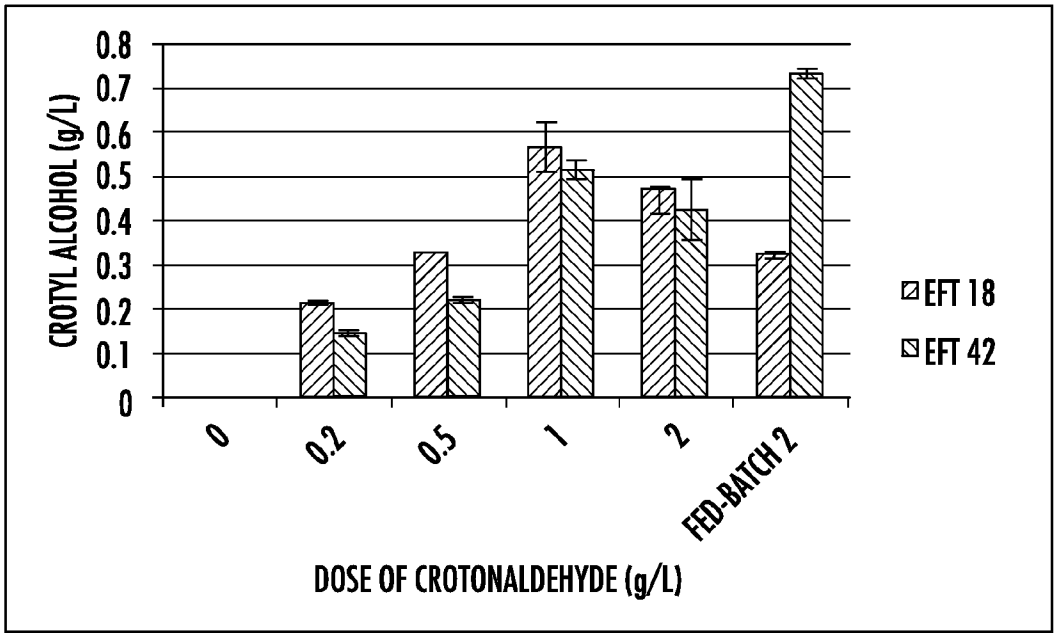


FIG. 24

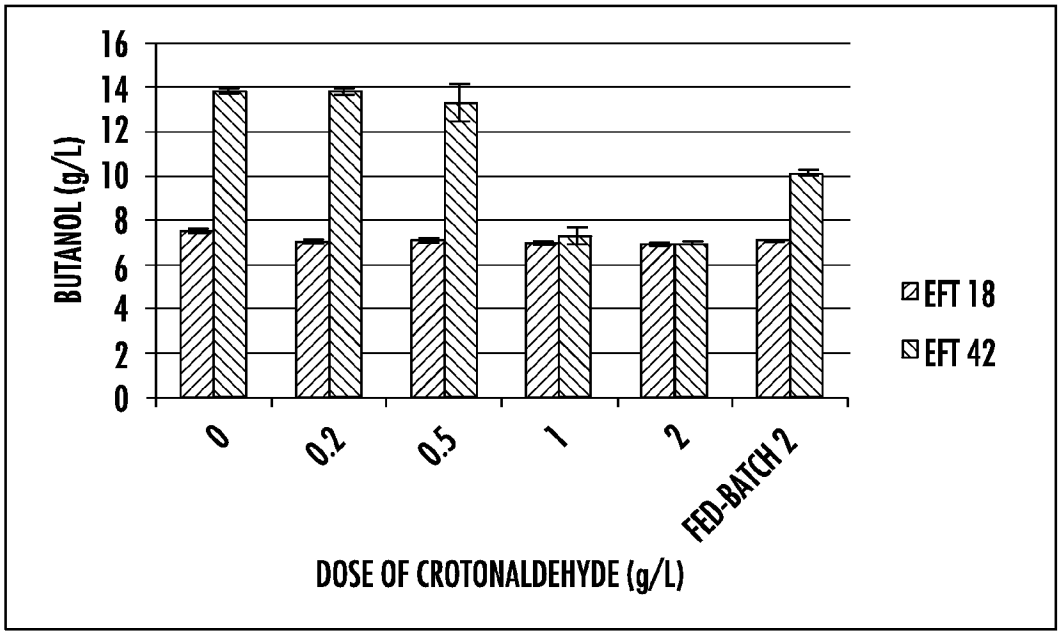


FIG. 25

9/13/2020, EAST Version: 4.1.7.1

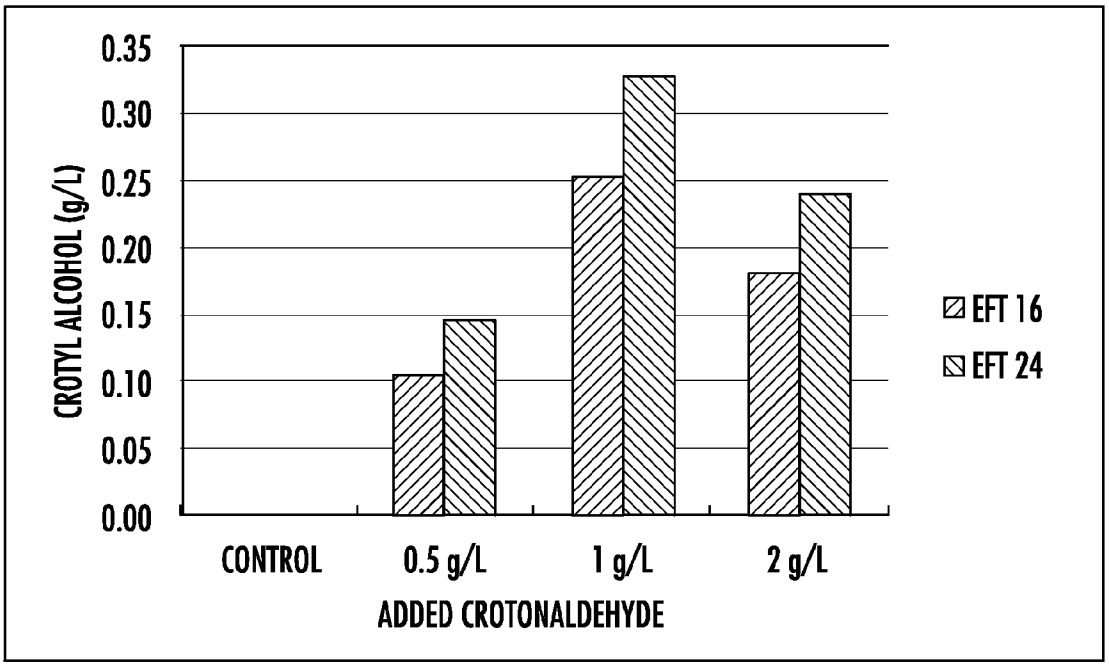


FIG. 26

9/13/2020, EAST Version: 4.1.7.1

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US 15/11371

A. CLASSIFICATION OF SUBJECT MATTER
IPC(8) - B01J 29/06, C07C 2/12, C07C 5/32, C07C 11/167 (2015.01)
CPC - B01J 29/06, C07C 2/12, C07C 5/32, C07C 5/321, C07C 11/167
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC(8)- B01J 29/06, C07C 2/12, C07C 5/32, C07C 11/167 (2015.01);
CPC- B01J 29/06, C07C 2/12, C07C 5/32, C07C 5/321, C07C 11/167

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
USPC- 208/187, 208/49, 208/DIG2, 585/252, 585/475, 585/481, 585/615, 585/664;
Patents and NPL (classification, keyword; search terms below)

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
Pub West (US EP JP WO), Pat Base (AU BE BR CA CH CN DE DK EP ES FI FR GB IN JP KR SE TH TW US WO), Google Patent, Google Scholar, Free Patents Online; search terms: dehydrate, dehydrogenate, hydroform, diene, butadiene, alkenol, diol, crotyl, alcohol, catalyst, inorganic, alumina, organosilane, alkoxysilane, chlorosilane, calcium, diesel, jet, fuel

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 2012/0225466 A1 (BURK et al.) 06 September 2012 (06.09.2012), para [0007], [0045], [0046], [0243]	1-31
Y	US 2007/0144939 A1 (ZONES et al.) 28 June 2007 (28.06.2007), para [0040], [0046], [0065]-[0068], [0102], [0114], [0116], [0139]	1-31
Y	US 5,498,811 A (PEREGO et al.) 12 March 1996 (12.03.1996), col 2, ln 20-30; col 3, ln 13-17; col 3, ln 43-61	14, 19, 23-28
Y	US 5,476,982 A (ROBBINS et al.) 19 December 1995 (19.12.1995), col 1, ln 35-50; col 2, ln 18-57; col 5, ln 9-46; col 12, ln 8-10	15-17
Y	US 3,835,199 A (SCHWARTZ JR et al.) 10 September 1974 (10.09.1974), col 1, ln 48 to col 6, ln 60	1-31
Y	US 2,407,291 A (QUATTLEBAUM et al.) 10 September 1946 (10.09.1946), col 2, ln 17 to col 6, ln 75	1-31

☐ Further documents are listed in the continuation of Box C. ☐

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" earlier application or patent but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
30 May 2015 (30.05.2015)	24 JUN 2015
Name and mailing address of the ISA/US Mail Stop PCT, Attn: ISA/US, Commissioner for Patents P.O. Box 1450, Alexandria, Virginia 22313-1450 Facsimile No. 571-273-8300	Authorized officer: Lee W. Young PCT Helpdesk: 571-272-4300 PCT OSP: 571-272-7774

Form PCT/ISA/210 (second sheet) (January 2015)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US 15/11371

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

-- Please See Extra Sheet --

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying additional fees, this Authority did not invite payment of additional fees.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

1-31

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.

☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.

☐ No protest accompanied the payment of additional search fees.

Form PCT/ISA/210 (continuation of first sheet (2)) (January 2015)

9/13/2020, EAST Version: 4.1.7.1

Continued from Box No. III, Observations where unity of invention is lacking,

The only feature shared by Groups I, III, IV, and V that would otherwise unify the groups is one or more Group 2 element(s). However, this shared technical feature does not represent a contribution over prior art, because the shared technical feature is anticipated by Zones.

Zones discloses one or more Group 2 element(s) (para [0114], Group II.).

The only feature shared by Groups I and V that would otherwise unify the groups is one or more buffer-adjusted anion(s) at a WHSV at least about 0.3 hr-1. However, this shared technical feature does not represent a contribution over prior art, because the shared technical feature is anticipated by Zones.

Zones discloses one or more buffer-adjusted anion(s) (para [0040], [0046]) at a WHSV at least about 0.3 hr-1 (para [0040], [0069], [0108], SSZ-74 catalysts comprise SDA dication for production of halide to hydroxide ions... superscript 5 WHSV, having values of about 0.1 per hour.)

The only feature shared by Groups III, IV, and V that would otherwise unify the groups is at least one inorganic support. However, this shared technical feature does not represent a contribution over prior art, because the shared technical feature is anticipated by Zones.

Zones discloses at least one inorganic support (para [0062], matrix materials such as clay, silica, metal oxides.).

The only feature shared by Groups V, VI, and VII that would otherwise unify the groups is crotyl alcohol. However, this shared technical feature does not represent a contribution over prior art, because the shared technical feature is anticipated by US 2013/0109064 A1 to Osterhout, et al. (hereinafter 'Osterhout').

Osterhout discloses crotyl alcohol (para [0012], [0025], [0105]).

As the technical feature was known in the art at the time of the invention, this cannot be considered a special technical feature that would otherwise unify the groups.

Groups I-VII therefore lack unity under PCT Rule 13 because they do not share a same or corresponding special technical feature.

Doc Code:IDS.3P
Document Description: Third-Party Submission Under 37 CFR 1.290

PTO/SB/429(08-12)
Approved for use through 07/31/2015. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

U.S. PATENTS

Cite No	Patent Number	Kind Code[1]	Issue Date (YYYY-MM-DD)	First Named Inventor
1	8912373	B2	2014-12-16	Michael Wright
2	9856461	B2	2018-01-02	Jeffrey Fortman

U.S. PATENT APPLICATION PUBLICATIONS

Cite No	Publication Number	Kind Code[1]	Publication Date (YYYY-MM-DD)	First Named Inventor

FOREIGN PATENTS AND PUBLISHED FOREIGN PATENT APPLICATIONS

Cite No	Foreign Document Number[3]	Country Code[2]	Kind Code[1]	Publication Date (YYYY-MM-DD)	Applicant, Patentee or First Named Inventor	T[5]
						☐

NON-PATENT PUBLICATIONS (e.g., journal article, Office action)

Cite No	Author (if any), title of the publication, page(s) being submitted, publication date, publisher (where available), place of publication (where available).	T[5]	E[6]

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

1	Klug et al., Degradation of Hydrocarbons by Members of the Genus Candida, II. Oxidation of n-Alkanes and 1-Alkenes by Candida lipolytica, Journal of Bacteriology, Vol. 93, No. 6, pp 1847-1852 (1967)	☐	☐
2	Liu et al., Hydrogen peroxide-independent production of α-alkenes by OleTJE P450 fatty acid decarboxylase, Biotechnology for Biofuels, 7:28 (2014)	☐	☐

STATEMENTS

The party making the submission is not an individual who has a duty to disclose information with respect to the above-identified application under 37 CFR 1.56.

This submission complies with the requirements of 35 U.S.C. 122(e) and 37 CFR 1.290.

☒ The fee set forth in 37 CFR 1.290(f) has been submitted herewith.

☐ The fee set forth in 37 CFR 1.290(f) is not required because this submission lists three or fewer total items and, to the knowledge of the person signing the statement after making reasonable inquiry, this submission is the first and the only submission under 35 U.S.C 122(e) filed in the above-identified application by the party making the submission or by a party in privity with the party.

☐ This resubmission is being made responsive to a notification of non-compliance issued for an earlier filed third-party submission. The corrections in this resubmission are limited to addressing the non-compliance. As such, the party making this resubmission: (1) requests that the Office apply the previously-paid fee set forth in 37 CFR 1.290(f), or (2) states that no fee is required to accompany this resubmission as the undersigned is again making the fee exemption statement set forth in 37 CFR 1.290(g).

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

Signature	/daniel altman/		
Name/Print	Daniel Altman	Registration Number (if applicable)	34115

Examiner Signature	/NANNETTE HOLLOMAN/	Date Considered	09/11/2020

*EXAMINER: Signature indicates all documents listed above have been considered,except for citations through which a line is drawn. Draw line through citation if not considered. Include a copy of this form with next communication to applicant. 1. If known, enter kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16. See MPEP 901.04(a). 2. Enter the country or patent office that issued the document, by two-letter code under WIPO standard ST.3. See MPEP 1851. 3. For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. 4. If known, enter the kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 . See MPEP 901.04(a). 5. Check mark indicates translation attached. 6. Check mark indicates evidence of publication attached.

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

PTO/SB/06 (09-11)
Approved for use through 1/31/2014. OMB 0651-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

PATENT APPLICATION FEE DETERMINATION RECORD Substitute for Form PTO-875	Application or Docket Number 16/262,902	Filing Date 01/30/2019	☐ To be Mailed

ENTITY: ☑ LARGE ☐ SMALL ☐ MICRO

APPLICATION AS FILED - PART I

FOR	(Column 1) NUMBER FILED	(Column 2) NUMBER EXTRA		RATE ($)	FEE ($)
☐ BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A		N/A	
☐ SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A		N/A	
☐ EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A		N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	minus 20 =	*		x $100 =	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	minus 3 =	*		x $460 =	
☐ APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $310 ($155 for small entity) for each additional 50 sheets or fraction thereof. See 35 U.S.C. 41(a)(1)(G) and 37 CFR 1.16(s).				
☐ MULTIPLE DEPENDENT CLAIM PRESENT (37 CFR 1.16(j))					
* If the difference in column 1 is less than zero, enter "0" in column 2.				TOTAL	

APPLICATION AS AMENDED - PART II

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT	01/19/2021	CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	* 23	Minus	** 23	= 0		x $100 =	0
	Independent (37 CFR 1.16(h))	* 2	Minus	*** 3	= 0		x $480 =	0
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	0

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT		CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	*	Minus	**	=		x $0 =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=		x $0 =	
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	

* If the entry in column 1 is less than the entry in column 2, write "0" in column 3.	LIE
** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 20, enter "20".	/TINA J BARDEN/
*** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 3, enter "3".	
The "Highest Number Previously Paid For" (Total or Independent) is the highest number found in the appropriate box in column 1.	

This collection of information is required by 37 CFR 1.16. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor: **Rocco V. Burgo**

Confirmation No.: **1028**

Application No.: **16/262,902**

Art Unit: **1612**

Filing Date: **January 30, 2019**

Examiner: **HOLLOMAN, Nannette**

Title: **Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same**

Attorney Docket No: **INLX.0198U2/484799**

VIA EFS

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

RESPONSE TO OFFICE ACTION UNDER 37 C.F.R. § 1.111

In response to the Non-Final Office Action mailed September 17, 2020, Applicants request reconsideration of this application in view of the following claim amendments and remarks:

A marked-up **Listing of the Claims** begin on page 2.

Remarks begin on page 6.

PTO/SB/22 (10-20)
Approved for use through 11/30/2020. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

PETITION FOR EXTENSION OF TIME UNDER 37 CFR 1.136(a)

Docket Number (Optional): INLX.0198U2/484799

Application Number: 16/262,902

Filed: January 30, 2019

For: Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same

Art Unit: 1612

Examiner: Nannette Holloman

This is a request under the provisions of 37 CFR 1.136(a) to extend the period for filing a reply in the above-identified application.

The requested extension and fee are as follows (check time period desired and enter the appropriate fee below):

	Fee	Small Entity Fee	Micro Entity Fee	
☑ One month (37 CFR 1.17(a)(1))	$220	$110	$55	$ 220.00
☐ Two months (37 CFR 1.17(a)(2))	$640	$320	$160	$
☐ Three months (37 CFR 1.17(a)(3))	$1,480	$740	$370	$
☐ Four months (37 CFR 1.17(a)(4))	$2,320	$1,160	$580	$
☐ Five months (37 CFR 1.17(a)(5))	$3,160	$1,580	$790	$

☐ Applicant asserts small entity status. See 37 CFR 1.27.

☐ Applicant certifies micro entity status. See 37 CFR 1.29.
Form PTO/SB/15A or B or equivalent must either be enclosed or have been submitted previously.

☐ A check in the amount of the fee is enclosed.

☐ Payment by credit card. Form PTO-2038 is attached.

☐ The Director has already been authorized to charge fees in this application to a Deposit Account.

☑ The Director is hereby authorized to charge any fees which may be required, or credit any overpayment, to Deposit Account Number 50-3111.

☑ Payment made via EFS-Web.

WARNING: Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.

I am the

☐ applicant/inventor.

☐ assignee of record of the entire interest. See 37 CFR 3.71. 37 CFR 3.73(b) statement is enclosed (Form PTO/SB/96).

☑ attorney or agent of record. Registration number 59,443.

☐ attorney or agent acting under 37 CFR 1.34. Registration number _____.

/Aaron Lukas, Reg.# 59,443/
Signature

January 19, 2021
Date

Aaron S. Lukas
Typed or printed name

202-912-4823
Telephone Number

NOTE: This form must be signed in accordance with 37 CFR 1.33. See 37 CFR 1.4 for signature requirements and certifications. Submit multiple forms if more than one signature is required, see below*.

☑ * Total of 1 forms are submitted.

This collection of information is required by 37 CFR 1.136(a). The information is required to obtain or retain a benefit by the public, which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.11 and 1.14. This collection is estimated to take 6 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

Privacy Act Statement

The **Privacy Act of 1974 (P.L. 93-579)** requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether disclosure of these records is required by the Freedom of Information Act.
2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.
3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.
4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).
5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.
6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).
7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (*i.e.*, GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.
8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspection or an issued patent.
9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

Electronic Patent Application Fee Transmittal

Application Number:	16262902
Filing Date:	30-Jan-2019
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Filer:	Aaron Scott Lukas/Mary Bertolino
Attorney Docket Number:	INLX.0198U2/484799

Filed as Large Entity

Filing Fees for Utility under 35 USC 111(a)

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Pages:				
Claims:				
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
Extension-of-Time:				

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Extension - 1 month with $0 paid	1251	1	220	220
Miscellaneous:				
			Total in USD ($)	**220**

Electronic Acknowledgement Receipt

EFS ID:	41678605
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	171052
Filer:	Aaron Scott Lukas/Mary Bertolino
Filer Authorized By:	Aaron Scott Lukas
Attorney Docket Number:	INLX.0198U2/484799
Receipt Date:	19-JAN-2021
Filing Date:	30-JAN-2019
Time Stamp:	13:09:16
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	DA
Payment was successfully received in RAM	$220
RAM confirmation Number	E20211ID09384512
Deposit Account	
Authorized User	

The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows:

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1		INLX_0198U2_484799-Response_to_09172020_Office_Action.PDF	227726 a8df97900e24842aeb2345bc9344bce89e58ec5e	yes	8

	Multipart Description/PDF files in .zip description		
	Document Description	**Start**	**End**
	Applicant Arguments/Remarks Made in an Amendment	6	8
	Claims	2	5
	Amendment/Req. Reconsideration-After Non-Final Reject	1	1

Warnings:					
Information:					
2	Extension of Time	INLX_0198U2_484799-One_Month_Extension_of_Time.pdf	169815 e84604ac4aa996d20468fe0e90624730b224abb9	no	2
Warnings:					
Information:					
3	Fee Worksheet (SB06)	fee-info.pdf	31018 4c41e904a58ca46a4a2a774a0f308cfa802f908c	no	2
Warnings:					
Information:					
		Total Files Size (in bytes):	428559		

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

REMARKS

Application and Claims Status

Upon entry of the foregoing claim amendments, claims 42-55 and 72-80 are pending, and claims 1-42 and 56-71 have been cancelled.

Claims 42-55 and 72-80 are presently amended. No new matter has been added. Support for the amendments to claims 42 and 72 can be found, *inter alia*, at ¶¶ [0038], [0061]-[0062], [0064] and [0089] of the as-filed specification. Support for the amendments to claims 47, 54, 55, 73-75,79, and 80 can be found, *inter alia*, at ¶¶ [0028]-[0029], [0064]-[0070], [0088], [0091], [0092], and FIG. 1 of the as-filed specification. Support for the amendments to claims 50 and 76 can be found, *inter alia*, at ¶¶ [0093] and [0105] of the as-filed specification. Support for the amendments to claims 51, 77, and 78 can be found, *inter alia*, at ¶ [0094], [0099], and [0118] of the as-filed specification. Support for the amendment to claim 53 can be found, *inter alia*, at ¶¶ [0028], [0046]-[0048], and [0050] of the as-filed specification.

Applicants note that the present claim amendments are made to advance the prosecution of the instant application. These amendments do not constitute any waiver or surrender of any subject matter that Applicants may be entitled to pursue, at their option, in a future submission such as a continuing or reissue application. The amendments similarly do not constitute any agreement or acquiescence as to the merits of the Office Action. Accordingly, Applicants expressly reserve the right to pursue the scope of any of the original, previously presented, withdrawn, or cancelled claims, as well as other claim scope, in a future submission.

Claims 42-55 and 72-80 are rejected. In view of the above claim amendments and following remarks, Applicants respectfully submit that the pending rejections have been traversed and respectfully request favorable reconsideration and allowance of the pending claims.

Patentability of the Claims

35 U.S.C. § 103, Obviousness—Claims 42-55 and 72-80

The Office has rejected claims 42-55 and 72-80 under 35 U.S.C. § 103 as allegedly obvious over Lutz (US Patent Pub. No. 2012/0282207) in view of Wang (*Green Chem.*, 14:758-65 (2012), and Wright (PCT Pub. No. WO 2016/007196).

Lutz discloses antimicrobial compositions that contain two or more "natural or naturally derived" fatty acids that can be mixed with other "natural or naturally derived products," an alcoholic solvent, and/or an organic acid. *See* Lutz, Abstract, ¶¶ [0006]-[0012]. The Office acknowledges that Lutz does not disclose the use of "bio" alcohols such as the presently claimed bio-1,2-alkanediols. Office Action at 3. The Office asserts that a person ordinary skill in the art would have relied on the disclosure of Wang to prepare bio-1,2-alkanediols for use in the compositions of Lutz. Office Action at 3. Finally, the Office asserts that a person of ordinary skill in the art would have relied on Wright in combination with Lutz and Wang to prepare bio-1,2-alkene intermediates having a regioselectivity of 90-99%, which impliedly, a person of ordinary skill in the art would have used to prepare the claimed compositions. Office Action at 4.

Applicants respectfully traverse this rejection.

The present independent claims 42 and 72 require "[a] preservative composition comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of about 8 to about 20 carbon atoms, wherein the carbon atoms of the bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived." Lutz admittedly fails to teach or suggest how to prepare bio-1,2-alkanediols. Wang discloses methods to prepare a wide range of compounds, including C_2-C_6 alkanediols, from cellulosic feedstocks. *See* Wang at 761, Table 1. However, Wang's processes are limited to the production of 1,2-alkanediols "with carbon number[s] in the range of 2-6." Wang at 759, col. 1. Conversely, the current claims require bio-1,2-alkanediols having a carbon chain length of about 8 to about 20 carbon atoms. A person of ordinary skill in the art following Wang would have no way to make any 1,2-alkanediols having a chain length longer than six carbon atoms because Wang's disclosure is limited to making 1,2-alkanediols based on ring-opening hydrogenolysis of cellulose. Wang, Abstract. Nor does Wang (or any other reference) teach or suggest ways that a person of ordinary skill in the art could have modified Wang's process to create longer-chain 1,2-alkanediols. As such, the presently claimed compositions would not have been obvious to a person of ordinary skill in the art over Lutz in view of Wang. For these reasons, Applicants respectfully request reconsideration and withdrawal of the rejection over Lutz in view of Wang.

Turning to Wright, the present claims do not recite a percent regioselectivity of a predecessor alkene. Notwithstanding this fact, the Office fails to explain how the alkene

compounds of Wright would have been relevant to the process of Wang, which uses cellulosic starting materials. Indeed, it is only through impermissible hindsight that a person of ordinary skill in the art would combine the teachings of Wright and Wang and/or Lutz. For these reasons, Applicants respectfully request reconsideration of any rejection involving Wright.

For the reasons stated above, Applicants respectfully request withdrawal the rejection of claims 42 and 72 as well as dependent claims 43-55 and 73-80.

CONCLUSION

In view of the foregoing remarks, it is respectfully submitted that each of the pending claims is in condition for allowance, and a Notice of Allowance is earnestly solicited. The Examiner is invited to contact the undersigned attorney with any questions, comments or suggestions relating to the referenced patent application.

This response is being submitted on January 19, 2021, with a petition for a one-month extension of time and associated fees. In the event the Commissioner determines that any additional fees are required for any reason, the Commissioner is hereby authorized to deduct said fee from Deposit Account No.: 50-3111/Matter No.: 484799.000.

Respectfully submitted,

Date: January 19, 2021

/Aaron Lukas, Reg.# 59,443/

Aaron S. Lukas, Ph.D.
Reg. No. 59,443
COZEN O'CONNOR
1200 Nineteenth Street N.W.
Washington, D.C. 20036
Tel: (202) 912-4823
Fax: (202) 618-4829

LISTING OF THE CLAIMS

This listing of claims will replace all prior versions, and listings, of claims in the application.

What is Claimed is:

1-41. (Canceled)

42. (Currently Amended) A preservative composition, comprising ~~at least one first~~ about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of about 8[[5]] to about 20 carbon atoms, wherein the carbon atoms of the first bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived~~which is synthesized by conversion of a first bio-alkene having a carbon chain of about 8[[5]] to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%~~.

43. (Currently Amended) A personal care composition comprising the [[The]]preservative composition of ~~according to~~ claim 42~~, wherein the composition is a personal care composition~~.

44. (Currently Amended) The personal care composition of ~~according to~~ claim 43, wherein the personal care composition is a hair care composition, an oral care composition, a skin care composition or a cosmetic composition.

45. (Currently Amended) The personal care composition of ~~according to~~ claim 43[[42]], wherein the personal care composition is ~~a composition for~~ a household product.

46. (Currently Amended) The personal care composition of ~~according to~~ claim 45, wherein the household product is a fabric care product or a cleaning product.

47. (Currently Amended) The personal care composition of ~~according to~~ claim 43[[42]], wherein the bio-1,2-alkanediol is selected from the group consisting of: bio-1,2-octanediol, bio-1,2-decanediol, bio-1,2-dodecanediol, and combinations thereof~~composition is an industrial composition~~.

48. (Currently Amended) The personal care composition of ~~according to~~ claim 43[[42]], wherein the personal care composition is a pharmaceutical composition, a vitamin composition, or a health care composition.

49. (Currently Amended) The personal care composition of ~~according to~~ claim 43[[42]], wherein the preservative composition is present in a concentration of about 0.1% to about 10%~~comprising a second bio-1,2-alkanediol, different than the first bio-1,2-alkanediol,~~

~~the second bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms which is synthesized by conversion of a second bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%~~.

50. (Currently Amended) The personal care composition of ~~according to~~ claim 43[[49]], ~~wherein the composition~~ further comprising ~~comprises at least one other~~ a bio-compound selected from the group consisting of: a bio-organic acid, bio-hydroxamic acid, a bio-alkylhydroxamic acid, a bio-terpenoid compound having 5 to 20 carbon atoms, bio-1,3-propanediol, bio-1,2-butanediol, bio-1,2-pentanediol, and combinations thereof~~different from the first and the second bio-1,2-alkanediols~~.

51. (Currently Amended) The personal care composition of ~~according to~~ claim 50, wherein the preservative composition is present in a concentration of about 0.1% to about 10%~~comprises an antimicrobial compound different from the first and the second bio-1,2-alkanediol and different from the other bio-compound~~.

52. (Currently Amended) The preservative composition of ~~according to~~ claim 42, wherein the composition further comprises at least one other bio-compound different from the ~~at least one~~ bio-1,2-alkanediol.

53. (Currently Amended) The preservative composition of ~~according to~~ claim 52, further comprising an ~~wherein the composition comprises at least one~~ antimicrobial compound different from the ~~at least one~~ bio-1,2-alkanediol ~~and different from the other bio-compound~~.

54. (Currently Amended) The preservative composition of ~~according to~~ claim 42, comprising about 75% to about 99% of ~~wherein~~ the bio-1,2-alkanediol~~first bio-1-alkene has a regioselectivity of about 95% to about 99%~~.

55. (Currently Amended) The preservative composition of ~~according to~~ claim 42, wherein the ~~first bio-alkene is bio-octene and the at least one first~~ bio-1,2-alkanediol comprises about 60% to about 99% [[is]]bio-1,2-octanediol.

56-71. (Canceled)

72. (Currently Amended) A preservative composition ~~An antimicrobial product,~~ comprising ~~at least one first~~ about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of about 8 [[5]]to about 14 [[20]]carbon atoms, wherein the carbon

atoms of the bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived~~which is synthesized by conversion of a first bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%~~.

73. (Currently Amended) The preservative composition of ~~antimicrobial product according to~~ claim 72, wherein the bio-1,2-alkanediol is selected from the group consisting of: bio-1,2-octanediol, bio-1,2-decanediol, bio-1,2-dodecanediol, and combinations thereof~~further comprising a second bio-1,2-alkanediol, different than the at least one first bio-1,2-alkanediol, the second bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms and formed by conversion of a second bio-alkene, different than the first bio-alkene, having a carbon chain length of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%~~.

74. (Currently Amended) The preservative composition of ~~antimicrobial product according to~~ claim 72[[73]], wherein the bio-1,2-alkanediol comprises about 60% to about 99% of bio-1,2-octanediol~~product further comprises at least one other bio-compound different from the at least one first and the second bio-1,2-alkanediols~~.

75. (Currently Amended) The preservative composition of ~~antimicrobial product according to~~ claim 72[[74]], wherein the bio-1,2-alkanediol comprises about 75% to about 99% bio-1,2-octanediol~~further comprising a second antimicrobial and/or a preservative different from the antimicrobial product~~.

76. (Currently Amended) The preservative composition of ~~antimicrobial product according to~~ claim 72, ~~wherein the product~~ further comprising a ~~comprises at least one other~~ bio-compound selected from the group consisting of: a bio-organic acid, bio-hydroxamic acid, a bio-alkylhydroxamic acid, a bio-terpenoid compound having 5 to 20 carbon atoms, bio-1,3-propanediol, bio-1,2-butanediol, bio-1,2-pentanediol, bio-1,2-hexanediol, and combinations thereof~~different from the at least one first bio-1,2-alkanediol~~.

77. (Currently Amended) A personal care composition comprising the preservative composition of ~~The antimicrobial product according to~~ claim 72 in a concentration of about 0.1% to about 10%~~76, further comprising a second antimicrobial and/or a preservative different from the antimicrobial product~~.

78. (Currently Amended) The personal care composition of ~~antimicrobial product according to~~ claim 77, wherein the preservative composition is present in a concentration of about 0.3% to about 2%~~72, wherein the first bio-1-alkene has a regioselectivity of about 95% to about 99%~~.

79. (Currently Amended) The personal care composition of ~~antimicrobial product according to~~ claim 78[[72]], wherein the preservative composition comprises about 60% to about 99% of bio-1,2-octanediol~~antimicrobial product demonstrates antimicrobial efficacy~~.

80. (Currently Amended) The personal care composition of ~~antimicrobial product according to~~ claim 78[[72]], wherein the preservative composition comprises about 75% to about 99% of ~~antimicrobial first bio-alkene is bio-octene and the at least one first 1,2-alkane dial is~~ bio-1,2-octanediol.

81-87. (Canceled)

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

NOTICE OF ALLOWANCE AND FEE(S) DUE

171052 7590 04/14/2021

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

EXAMINER
HOLLOMAN, NANNETTE

ART UNIT	PAPER NUMBER
1612	

DATE MAILED: 04/14/2021

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	INLX.0198U2/484799	1028

TITLE OF INVENTION: Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same

APPLN. TYPE	ENTITY STATUS	ISSUE FEE DUE	PUBLICATION FEE DUE	PREV. PAID ISSUE FEE	TOTAL FEE(S) DUE	DATE DUE
nonprovisional	UNDISCOUNTED	$1200	$0.00	$0.00	$1200	07/14/2021

THE APPLICATION IDENTIFIED ABOVE HAS BEEN EXAMINED AND IS ALLOWED FOR ISSUANCE AS A PATENT. PROSECUTION ON THE MERITS IS CLOSED. THIS NOTICE OF ALLOWANCE IS NOT A GRANT OF PATENT RIGHTS. THIS APPLICATION IS SUBJECT TO WITHDRAWAL FROM ISSUE AT THE INITIATIVE OF THE OFFICE OR UPON PETITION BY THE APPLICANT. SEE 37 CFR 1.313 AND MPEP 1308.

THE ISSUE FEE AND PUBLICATION FEE (IF REQUIRED) MUST BE PAID WITHIN THREE MONTHS FROM THE MAILING DATE OF THIS NOTICE OR THIS APPLICATION SHALL BE REGARDED AS ABANDONED. THIS STATUTORY PERIOD CANNOT BE EXTENDED. SEE 35 U.S.C. 151. THE ISSUE FEE DUE INDICATED ABOVE DOES NOT REFLECT A CREDIT FOR ANY PREVIOUSLY PAID ISSUE FEE IN THIS APPLICATION. IF AN ISSUE FEE HAS PREVIOUSLY BEEN PAID IN THIS APPLICATION (AS SHOWN ABOVE), THE RETURN OF PART B OF THIS FORM WILL BE CONSIDERED A REQUEST TO REAPPLY THE PREVIOUSLY PAID ISSUE FEE TOWARD THE ISSUE FEE NOW DUE.

HOW TO REPLY TO THIS NOTICE:

I. Review the ENTITY STATUS shown above. If the ENTITY STATUS is shown as SMALL or MICRO, verify whether entitlement to that entity status still applies.

If the ENTITY STATUS is the same as shown above, pay the TOTAL FEE(S) DUE shown above.

If the ENTITY STATUS is changed from that shown above, on PART B - FEE(S) TRANSMITTAL, complete section number 5 titled "Change in Entity Status (from status indicated above)".

For purposes of this notice, small entity fees are 1/2 the amount of undiscounted fees, and micro entity fees are 1/2 the amount of small entity fees.

II. PART B - FEE(S) TRANSMITTAL, or its equivalent, must be completed and returned to the United States Patent and Trademark Office (USPTO) with your ISSUE FEE and PUBLICATION FEE (if required). If you are charging the fee(s) to your deposit account, section "4b" of Part B - Fee(s) Transmittal should be completed and an extra copy of the form should be submitted. If an equivalent of Part B is filed, a request to reapply a previously paid issue fee must be clearly made, and delays in processing may occur due to the difficulty in recognizing the paper as an equivalent of Part B.

III. All communications regarding this application must give the application number. Please direct all communications prior to issuance to Mail Stop ISSUE FEE unless advised to the contrary.

IMPORTANT REMINDER: Maintenance fees are due in utility patents issuing on applications filed on or after Dec. 12, 1980. It is patentee's responsibility to ensure timely payment of maintenance fees when due. More information is available at www.uspto.gov/PatentMaintenanceFees.

PART B - FEE(S) TRANSMITTAL

Complete and send this form, together with applicable fee(s), by mail or fax, or via EFS-Web.

By mail, send to: Mail Stop ISSUE FEE
Commissioner for Patents
P.O. Box 1450
Alexandria, Virginia 22313-1450

By fax, send to: (571)-273-2885

INSTRUCTIONS: This form should be used for transmitting the ISSUE FEE and PUBLICATION FEE (if required). Blocks 1 through 5 should be completed where appropriate. All further correspondence including the Patent, advance orders and notification of maintenance fees will be mailed to the current correspondence address as indicated unless corrected below or directed otherwise in Block 1, by (a) specifying a new correspondence address; and/or (b) indicating a separate "FEE ADDRESS" for maintenance fee notifications.

CURRENT CORRESPONDENCE ADDRESS (Note: Use Block 1 for any change of address)

171052 7590 04/14/2021

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

Note: A certificate of mailing can only be used for domestic mailings of the Fee(s) Transmittal. This certificate cannot be used for any other accompanying papers. Each additional paper, such as an assignment or formal drawing, must have its own certificate of mailing or transmission.

Certificate of Mailing or Transmission

I hereby certify that this Fee(s) Transmittal is being deposited with the United States Postal Service with sufficient postage for first class mail in an envelope addressed to the Mail Stop ISSUE FEE address above, or being transmitted to the USPTO via EFS-Web or by facsimile to (571) 273-2885, on the date below.

(Typed or printed name)
(Signature)
(Date)

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	INLX.0198U2/484799	1028

TITLE OF INVENTION: Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same

APPLN. TYPE	ENTITY STATUS	ISSUE FEE DUE	PUBLICATION FEE DUE	PREV. PAID ISSUE FEE	TOTAL FEE(S) DUE	DATE DUE
nonprovisional	UNDISCOUNTED	$1200	$0.00	$0.00	$1200	07/14/2021

EXAMINER	ART UNIT	CLASS-SUBCLASS
HOLLOMAN, NANNETTE	1612	424-049000

1. Change of correspondence address or indication of "Fee Address" (37 CFR 1.363).

❑ Change of correspondence address (or Change of Correspondence Address form PTO/SB/122) attached.

❑ "Fee Address" indication (or "Fee Address" Indication form PTO/SB/47; Rev 03-09 or more recent) attached. **Use of a Customer Number is required.**

2. For printing on the patent front page, list
(1) The names of up to 3 registered patent attorneys or agents OR, alternatively,
(2) The name of a single firm (having as a member a registered attorney or agent) and the names of up to 2 registered patent attorneys or agents. If no name is listed, no name will be printed.

1 _____
2 _____
3 _____

3. ASSIGNEE NAME AND RESIDENCE DATA TO BE PRINTED ON THE PATENT (print or type)

PLEASE NOTE: Unless an assignee is identified below, no assignee data will appear on the patent. If an assignee is identified below, the document must have been previously recorded, or filed for recordation, as set forth in 37 CFR 3.11 and 37 CFR 3.81(a). Completion of this form is NOT a substitute for filing an assignment.

(A) NAME OF ASSIGNEE (B) RESIDENCE: (CITY and STATE OR COUNTRY)

Please check the appropriate assignee category or categories (will not be printed on the patent) : ❑ Individual ❑ Corporation or other private group entity ❑ Government

4a. Fees submitted: ❑ Issue Fee ❑ Publication Fee (if required) ❑ Advance Order - # of Copies _____

4b. Method of Payment: *(Please first reapply any previously paid fee shown above)*

❑ Electronic Payment via EFS-Web ❑ Enclosed check ❑ Non-electronic payment by credit card (Attach form PTO-2038)

❑ The Director is hereby authorized to charge the required fee(s), any deficiency, or credit any overpayment to Deposit Account No. _____

5. **Change in Entity Status** (from status indicated above)

❑ Applicant certifying micro entity status. See 37 CFR 1.29

❑ Applicant asserting small entity status. See 37 CFR 1.27

❑ Applicant changing to regular undiscounted fee status.

NOTE: Absent a valid certification of Micro Entity Status (see forms PTO/SB/15A and 15B), issue fee payment in the micro entity amount will not be accepted at the risk of application abandonment.
NOTE: If the application was previously under micro entity status, checking this box will be taken to be a notification of loss of entitlement to micro entity status.
NOTE: Checking this box will be taken to be a notification of loss of entitlement to small or micro entity status, as applicable.

NOTE: This form must be signed in accordance with 37 CFR 1.31 and 1.33. See 37 CFR 1.4 for signature requirements and certifications.

Authorized Signature _____ Date _____

Typed or printed name _____ Registration No. _____

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	INLX.0198U2/484799	1028

171052 7590 04/14/2021

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

EXAMINER	
HOLLOMAN, NANNETTE	
ART UNIT	PAPER NUMBER
1612	

DATE MAILED: 04/14/2021

Determination of Patent Term Adjustment under 35 U.S.C. 154 (b)
(Applications filed on or after May 29, 2000)

The Office has discontinued providing a Patent Term Adjustment (PTA) calculation with the Notice of Allowance.

Section 1(h)(2) of the AIA Technical Corrections Act amended 35 U.S.C. 154(b)(3)(B)(i) to eliminate the requirement that the Office provide a patent term adjustment determination with the notice of allowance. See Revisions to Patent Term Adjustment, 78 Fed. Reg. 19416, 19417 (Apr. 1, 2013). Therefore, the Office is no longer providing an initial patent term adjustment determination with the notice of allowance. The Office will continue to provide a patent term adjustment determination with the Issue Notification Letter that is mailed to applicant approximately three weeks prior to the issue date of the patent, and will include the patent term adjustment on the patent. Any request for reconsideration of the patent term adjustment determination (or reinstatement of patent term adjustment) should follow the process outlined in 37 CFR 1.705.

Any questions regarding the Patent Term Extension or Adjustment determination should be directed to the Office of Patent Legal Administration at (571)-272-7702. Questions relating to issue and publication fee payments should be directed to the Customer Service Center of the Office of Patent Publication at 1-(888)-786-0101 or (571)-272-4200.

OMB Clearance and PRA Burden Statement for PTOL-85 Part B

The Paperwork Reduction Act (PRA) of 1995 requires Federal agencies to obtain Office of Management and Budget approval before requesting most types of information from the public. When OMB approves an agency request to collect information from the public, OMB (i) provides a valid OMB Control Number and expiration date for the agency to display on the instrument that will be used to collect the information and (ii) requires the agency to inform the public about the OMB Control Number's legal significance in accordance with 5 CFR 1320.5(b).

The information collected by PTOL-85 Part B is required by 37 CFR 1.311. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 30 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, Virginia 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, Virginia 22313-1450. Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

Privacy Act Statement

The Privacy Act of 1974 (P.L. 93-579) requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether disclosure of these records is required by the Freedom of Information Act.
2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.
3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.
4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).
5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.
6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).
7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.
8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspection or an issued patent.
9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

Notice of Allowability	Application No. 16/262,902	Applicant(s) Burgo et al.	
	Examiner NANNETTE HOLLOMAN	Art Unit 1612	AIA (FITF) Status Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address--

All claims being allowable, PROSECUTION ON THE MERITS IS (OR REMAINS) CLOSED in this application. If not included herewith (or previously mailed), a Notice of Allowance (PTOL-85) or other appropriate communication will be mailed in due course. **THIS NOTICE OF ALLOWABILITY IS NOT A GRANT OF PATENT RIGHTS.** This application is subject to withdrawal from issue at the initiative of the Office or upon petition by the applicant. See 37 CFR 1.313 and MPEP 1308.

1. ☑ This communication is responsive to arguments/amendments filed 01/19/2021.
   ☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____.

2. ☐ An election was made by the applicant in response to a restriction requirement set forth during the interview on _____; the restriction requirement and election have been incorporated into this action.

3. ☑ The allowed claim(s) is/are 42-55 and 72-80 . As a result of the allowed claim(s), you may be eligible to benefit from the **Patent Prosecution Highway** program at a participating intellectual property office for the corresponding application. For more information , please see **http://www.uspto.gov/patents/init_events/pph/index.jsp** or send an inquiry to **PPHfeedback@uspto.gov.**

4. ☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

   Certified copies:

   a) ☐ All b) ☐ Some *c) ☐ None of the:

   1. ☐ Certified copies of the priority documents have been received.
   2. ☐ Certified copies of the priority documents have been received in Application No. _____ .
   3. ☐ Copies of the certified copies of the priority documents have been received in this national stage application from the International Bureau (PCT Rule 17.2(a)).

   * Certified copies not received: _____ .

Applicant has THREE MONTHS FROM THE "MAILING DATE" of this communication to file a reply complying with the requirements noted below. Failure to timely comply will result in ABANDONMENT of this application.
THIS THREE-MONTH PERIOD IS NOT EXTENDABLE.

5. ☐ CORRECTED DRAWINGS (as "replacement sheets") must be submitted.
   ☐ including changes required by the attached Examiner's Amendment / Comment or in the Office action of Paper No./Mail Date _____ .

   Identifying indicia such as the application number (see 37 CFR 1.84(c)) should be written on the drawings in the front (not the back) of each sheet. Replacement sheet(s) should be labeled as such in the header according to 37 CFR 1.121(d).

6. ☐ DEPOSIT OF and/or INFORMATION about the deposit of BIOLOGICAL MATERIAL must be submitted. Note the attached Examiner's comment regarding REQUIREMENT FOR THE DEPOSIT OF BIOLOGICAL MATERIAL.

Attachment(s)

1. ☐ Notice of References Cited (PTO-892)
2. ☐ Information Disclosure Statements (PTO/SB/08), Paper No./Mail Date _____.
3. ☐ Examiner's Comment Regarding Requirement for Deposit of Biological Material _____.
4. ☐ Interview Summary (PTO-413), Paper No./Mail Date. _____.
5. ☐ Examiner's Amendment/Comment
6. ☐ Examiner's Statement of Reasons for Allowance
7. ☐ Other _____.

/NANNETTE HOLLOMAN/
Primary Examiner, Art Unit 1612

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

CPC - Searched*		
Symbol	**Date**	**Examiner**
A01N	04/10/2021	NH
C07C	04/10/2021	NH
A61K	04/10/2021	NH
A61Q	04/10/2021	NH

CPC Combination Sets - Searched*		
Symbol	**Date**	**Examiner**

US Classification - Searched*			
Class	**Subclass**	**Date**	**Examiner**

* See search history printout included with this form or the SEARCH NOTES box below to determine the scope of the search.

Search Notes		
Search Notes	**Date**	**Examiner**
PALM Inventor Search	09/13/2020	NH
EAST Search	09/13/2020	NH
NPL Search	09/13/2020	NH

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

Interference Search			
US Class/CPC Symbol	**US Subclass/CPC Group**	**Date**	**Examiner**
C07C	11/18, 2527/054, 2527/167, 2527/173, 2531/02, 65/21,	04/10/2021	NH
A61K	8/345, 2300/00, 2800/31, 8/60, 2800/412, 8/11, 8/732, 8/738, 31/197, 8/44, 8/73, 2800/48, 8/25, 8/34, 8/585, 8/92, 8/9789, 9/014, 2800/30, 31/198, 36/28, 36/534, 36/67	04/10/2021	NH
A61Q	15/00, 19/00, 13/00, 11/00, 5/00, 19/007, 19/08, 19/02, 17/005, 17/04, 19/10, 5/02, 7/00	04/10/2021	NH

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

CPC						
Symbol					Type	Version
C07C	/	29	/	04	F	2013-01-01
C07C	/	31	/	20	I	2013-01-01
C07C	/	29	/	80	I	2013-01-01
B01J	/	23	/	06	I	2013-01-01
B01J	/	37	/	08	I	2013-01-01
B01J	/	21	/	04	I	2013-01-01
C11D	/	3	/	2044	I	2013-01-01
C07C	/	1	/	24	I	2013-01-01
C07C	/	29	/	48	I	2013-01-01
C07C	/	11	/	02	I	2013-01-01
C07C	/	11	/	107	I	2013-01-01
B01J	/	2523	/	27	A	2013-01-01
C07B	/	41	/	02	A	2013-01-01
C07B	/	63	/	00	A	2013-01-01
B01J	/	2523	/	31	A	2013-01-01
A61L	/	2	/	18	A	2013-01-01
A01N	/	31	/	02	A	2013-01-01
A61K	/	8	/	345	A	2013-01-01
A61Q	/	5	/	00	A	2013-01-01
A61Q	/	11	/	00	A	2013-01-01
A61Q	/	19	/	00	A	2013-01-01
C11D	/	3	/	48	A	2013-01-01
C07C	/	2521	/	04	A	2013-01-01

CPC Combination Sets								
Symbol					Type	Set	Ranking	Version
C07C	/	1	/	24	I	1	1	2013-01-01
C07C	/	11	/	02	I	1	2	2013-01-01
C07C	/	29	/	48	I	2	1	2013-01-01
C07C	/	31	/	20	I	2	2	2013-01-01

NONE		Total Claims Allowed:	
(Assistant Examiner)	(Date)	23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	10 April 2021	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	NONE

U.S. Patent and Trademark Office

Part of Paper No.: 20210410

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	**Art Unit**
	NANNETTE HOLLOMAN	1612

CPC Combination Sets								
Symbol					**Type**	**Set**	**Ranking**	**Version**
C07C	/	29	/	80	I	3	1	2013-01-01
C07C	/	31	/	20	I	3	2	2013-01-01

NONE		**Total Claims Allowed:**	
(Assistant Examiner)	(Date)	23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	10 April 2021	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	NONE

U.S. Patent and Trademark Office

Part of Paper No.: 20210410

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

INTERNATIONAL CLASSIFICATION					
CLAIMED					
C07C29/04	/	29	/	04	

NON-CLAIMED				
	/		/	

US ORIGINAL CLASSIFICATION	
CLASS	SUBCLASS

CROSS REFERENCES(S)						
CLASS	SUBCLASS (ONE SUBCLASS PER BLOCK)					

NONE		Total Claims Allowed:	
(Assistant Examiner)	(Date)	23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	10 April 2021	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	NONE

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

☐ Claims renumbered in the same order as presented by applicant ☐ CPA ☐ T.D. ☐ R.1.47

CLAIMS

Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original
	1		11		21		31		41	10	51		61		71
	2		12		22		32	1	42	11	52		62	15	72
	3		13		23		33	2	43	12	53		63	16	73
	4		14		24		34	3	44	13	54		64	17	74
	5		15		25		35	4	45	14	55		65	18	75
	6		16		26		36	5	46		56		66	19	76
	7		17		27		37	6	47		57		67	20	77
	8		18		28		38	7	48		58		68	21	78
	9		19		29		39	8	49		59		69	22	79
	10		20		30		40	9	50		60		70	23	80

NONE		Total Claims Allowed:	
(Assistant Examiner)	(Date)	23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	10 April 2021	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	NONE

U.S. Patent and Trademark Office

Part of Paper No.: 20210410

Web Search History

date, time	web site	search string
4/10/2021 8:03:19 PM	Google Scholar	bio octanediol
4/10/2021 8:03:41 PM	Google Scholar	bio 1,2-octanediol

EAST Search History

EAST Search History (Prior Art)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
L1	67	"bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol)))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/04/10 19:56
L2	549,665	((A01N47/44 OR A01N37/36 OR A01N59/16 OR A01N31/02 OR A01N25/34 OR A01N59/00 OR A01N61/00 OR A01N25/00 OR A01N2300/00 OR A01N25/10 OR C07C31/20 OR C07C1/24 OR C07C11/02 OR C07C29/48 OR C07C29/80 OR C07C263/12 OR C07C265/14 OR C07C2521/04 OR C07C11/107 OR C07C29/04 OR C07C247/24 OR C07C11/167 OR C07C11/18 OR C07C2527/054 OR C07C2527/167 OR C07C2527/173 OR C07C2531/02 OR C07C65/21 OR A61K8/345 OR A61K2300/00 OR A61K2800/31 OR A61K8/60 OR A61K2800/412 OR A61K8/11 OR A61K8/732 OR A61K8/738 OR A61K31/197 OR A61K8/44 OR A61K8/73 OR A61K2800/48 OR A61K8/25 OR A61K8/34 OR A61K8/585 OR A61K8/92 OR A61K8/9789 OR A61K9/0014 OR A61K2800/30 OR A61K31/198 OR A61K36/28 OR A61K36/534 OR A61K36/67 OR A61K36/886 OR A61K45/06 OR A61K47/10 OR A61K8/24 OR A61K8/362 OR A61K8/37 OR A61K8/41 OR A61K8/42 OR A61K8/4973 OR A61K8/498 OR A61K8/891 OR A61K9/06 OR A61K2800/10 OR A61K2800/28 OR A61K2800/524 OR A61K2800/56 OR A61K2800/87 OR A61K2800/874 OR A61K31/327 OR A61K47/22 OR A61K47/44 OR A61K47/46 OR A61K8/0216 OR A61K8/046 OR A61K8/19 OR A61K8/20 OR A61K8/23 OR A61K8/26 OR A61K8/31 OR A61K8/64 OR A61K8/678 OR A61K8/735 OR A61K8/8111 OR A61K8/8147 OR A61K8/893 OR A61K8/895 OR	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/04/10 19:57

		A61K8/922 OR A61K8/9794 OR A61K9/107 OR A61K2035/115 OR A61K2800/43 OR A61K2800/522 OR A61K2800/592 OR A61K2800/5922 OR A61K2800/594 OR A61K2800/596 OR A61K2800/624 OR A61K2800/805 OR A61K31/192 OR A61K31/355 OR A61K31/60 OR A61K33/30 OR A61K35/745 OR A61K35/747 OR A61K36/185 OR A61K36/53 OR A61K47/02 OR A61K47/06 OR A61K47/12 OR A61K47/14 OR A61K47/183 OR A61K47/24 OR A61K47/34 OR A61K47/38 OR A61K49/0006 OR A61K49/0008 OR A61K8/06 OR A61K8/062 OR A61K8/28 OR A61K8/29 OR A61K8/342 OR A61K8/347 OR A61K8/365 OR A61K8/368 OR A61K8/375 OR A61K8/416 OR A61K8/445 OR A61K8/463 OR A61K8/49 OR A61K8/4926 OR A61K8/63 OR A61K8/66 OR A61K8/675 OR A61K8/8152 OR A61K8/8194 OR A61K8/927 OR A61K8/965 OR A61K8/97 OR A61K9/08 OR A61Q15/00 OR A61Q19/00 OR A61Q13/00 OR A61Q11/00 OR A61Q5/00 OR A61Q19/007 OR A61Q19/08 OR A61Q19/02 OR A61Q17/005 OR A61Q17/04 OR A61Q19/10 OR A61Q5/02 OR A61Q7/00).CPC.)				
L3	42	l2 and ("bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/04/10 19:57
S1	937	((("BURGO") near3 ("Rocco")) OR (("WRIGHT") near3 ("Michael")) OR (("MOSSER") near3 ("Gary")) OR (("FEVOLA") near3 ("Michael"))).INV.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	OFF	2020/07/07 19:12
S2	378	(bio with alkene) bioalkene	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/07/07 19:19
S3	6,577	regioselectivity	US-PGPUB; USPAT; USOCR;	OR	ON	2020/09/09 14:30

			EPO; JPO; DERWENT			
S4	13	regioselectivity same alkanediol	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/09 14:30
S5	7	regioselectivity same (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/09 14:47
S6	1	("7385092").PN.	USPAT; USOCR	OR	OFF	2020/09/09 14:53
S7	2	wo-2016007196-$.did.	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	OFF	2020/09/11 11:19
S8	6	alkanediol same (household and pharmaceutical and cosmetic)	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:25
S9	3	(household and pharmaceutical and cosmetic) same (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26
S10	336	(household and pharmaceutical and cosmetic) and (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26
S11	6	((household and pharmaceutical and cosmetic) and (octanediol or (caprylyl adj glycol))).clm.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26
S12	11	((household and pharmaceutical and cosmetic) and (octanediol or ((caprylyl or hexylene or decylene) adj glycol) or alkanediol)).clm.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:47
S13	946	((("BURGO") near3 ("Rocco")) OR (("WRIGHT") near3 ("Michael")) OR (("MOSSER") near3 ("Gary")) OR (("FEVOLA") near3 ("Michael"))).INV.	US-PGPUB; USPAT; USOCR;	OR	OFF	2020/09/11 19:14

			EPO; JPO; DERWENT			
S14	3	S13 and alkanediol	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/09/11 19:18
S15	7	(("8912373") or ("9856461") or ("4234752") or ("20050065389") or ("20030065233") or ("20060161025") or ("20140066666")).PN.	US-PGPUB; USPAT; USOCR	OR	OFF	2020/09/13 13:37
S16	42,261	decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/02/27 23:31
S17	13,209	S16 and (preservative or pharmaceutical or cosmetic)	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/02/27 23:32
S18	1,488	S17 and "bio"	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/02/27 23:32

EAST Search History (Interference)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
L4	125,400	((A01N47/44 OR A01N37/36 OR A01N59/16 OR A01N31/02 OR A01N25/34 OR A01N59/00 OR A01N61/00 OR A01N25/00 OR A01N2300/00 OR A01N25/10 OR C07C31/20 OR C07C1/24 OR C07C11/02 OR C07C29/48 OR C07C29/80 OR C07C263/12 OR C07C265/14 OR C07C2521/04 OR C07C11/107 OR C07C29/04 OR C07C247/24 OR C07C11/167 OR C07C11/18 OR C07C2527/054 OR C07C2527/167 OR C07C2527/173 OR C07C2531/02 OR C07C65/21 OR A61K8/345 OR A61K2300/00 OR A61K2800/31 OR A61K8/60 OR A61K2800/412 OR A61K8/11 OR A61K8/732 OR A61K8/738 OR A61K31/197 OR A61K8/44 OR A61K8/73 OR A61K2800/48 OR A61K8/25 OR A61K8/34 OR A61K8/585 OR A61K8/92 OR A61K8/9789 OR A61K9/0014 OR	USPAT	OR	ON	2021/04/10 19:59

		A61K2800/30 OR A61K31/198 OR A61K36/28 OR A61K36/534 OR A61K36/67 OR A61K36/886 OR A61K45/06 OR A61K47/10 OR A61K8/24 OR A61K8/362 OR A61K8/37 OR A61K8/41 OR A61K8/42 OR A61K8/4973 OR A61K8/498 OR A61K8/891 OR A61K9/06 OR A61K2800/10 OR A61K2800/28 OR A61K2800/524 OR A61K2800/56 OR A61K2800/87 OR A61K2800/874 OR A61K31/327 OR A61K47/22 OR A61K47/44 OR A61K47/46 OR A61K8/0216 OR A61K8/046 OR A61K8/19 OR A61K8/20 OR A61K8/23 OR A61K8/26 OR A61K8/31 OR A61K8/64 OR A61K8/678 OR A61K8/735 OR A61K8/8111 OR A61K8/8147 OR A61K8/893 OR A61K8/895 OR A61K8/922 OR A61K8/9794 OR A61K9/107 OR A61K2035/115 OR A61K2800/43 OR A61K2800/522 OR A61K2800/592 OR A61K2800/5922 OR A61K2800/594 OR A61K2800/596 OR A61K2800/624 OR A61K2800/805 OR A61K31/192 OR A61K31/355 OR A61K31/60 OR A61K33/30 OR A61K35/745 OR A61K35/747 OR A61K36/185 OR A61K36/53 OR A61K47/02 OR A61K47/06 OR A61K47/12 OR A61K47/14 OR A61K47/183 OR A61K47/24 OR A61K47/34 OR A61K47/38 OR A61K49/0006 OR A61K49/0008 OR A61K8/06 OR A61K8/062 OR A61K8/28 OR A61K8/29 OR A61K8/342 OR A61K8/347 OR A61K8/365 OR A61K8/368 OR A61K8/375 OR A61K8/416 OR A61K8/445 OR A61K8/463 OR A61K8/49 OR A61K8/4926 OR A61K8/63 OR A61K8/66 OR A61K8/675 OR A61K8/8152 OR A61K8/8194 OR A61K8/927 OR A61K8/965 OR A61K8/97 OR A61K9/08 OR A61Q15/00 OR A61Q19/00 OR A61Q13/00 OR A61Q11/00 OR A61Q5/00 OR A61Q19/007 OR A61Q19/08 OR A61Q19/02 OR A61Q17/005 OR A61Q17/04 OR A61Q19/10 OR A61Q5/02 OR A61Q7/00).CPC.)				
L5	10	l4 and ("bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))))	USPAT	OR	ON	2021/04/10 19:59

4/10/2021 8:04:42 PM
C:\Users\nholloman\Documents\EAST\Workspaces\16262902.wsp

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor: **Rocco V. Burgo**	Attorney Docket No: **INLX.0198U2/484799**
Application No.: **16/262,902**	Confirmation No.: **1028**
Filing Date: **January 20, 2019**	Art Unit: **1612**
Title: ***Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same***	Examiner: **HOLLOMAN, Nannette**

VIA EFS

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

AMENDMENT AFTER NOTICE OF ALLOWANCE UNDER 37 C.F.R. § 1.312

In response to the Notice of Allowability mailed April 14, 2021, Applicant files these papers under 37 C.F.R. § 1.312 and requests reconsideration of this application in view of the following claim amendments and remarks:

A marked-up **Listing of the Claims** begin on page 2.

Remarks begin on page 5.

LISTING OF THE CLAIMS

This listing of claims will replace all prior versions, and listings, of claims in the application.

What is claimed is:

1-41. (Canceled).

42. (Currently Amended) A preservative composition, comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of 7[[about 8]] to [[about]]20 carbon atoms, wherein the carbon atoms of the first bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived.

43. (Previously Presented) A personal care composition comprising the preservative composition of claim 42.

44. (Previously Presented) The personal care composition of claim 43, wherein the personal care composition is a hair care composition, an oral care composition, a skin care composition or a cosmetic composition.

45. (Previously Presented) The personal care composition of claim 43, wherein the personal care composition is a household product.

46. (Previously Presented) The personal care composition of claim 45, wherein the household product is a fabric care product or a cleaning product.

47. (Currently Amended) The personal care composition of claim 43, wherein the bio-1,2-alkanediol is selected from the group consisting of: bio-1,2-heptanediol, bio-1,2-octanediol, bio-1,2-decanediol, bio-1,2-dodecanediol, and combinations thereof.

48. (Previously Presented) The personal care composition of claim 43, wherein the personal care composition is a pharmaceutical composition, a vitamin composition, or a health care composition.

49. (Previously Presented) The personal care composition of claim 43, wherein the preservative composition is present in a concentration of about 0.1% to about 10%.

50. (Previously Presented) The personal care composition of claim 43, further comprising a bio-compound selected from the group consisting of: a bio-organic acid, bio-hydroxamic acid, a bio-alkylhydroxamic acid, a bio-terpenoid compound having 5 to 20 carbon atoms, bio-1,3-propanediol, bio-1,2-butanediol, bio-1,2-pentanediol, and combinations thereof.

51. (Previously Presented) The personal care composition of claim 50, wherein the preservative composition is present in a concentration of about 0.1% to about 10%.
52. (Previously Presented) The preservative composition of claim 42, wherein the composition further comprises at least one other bio-compound different from the bio-1,2-alkanediol.
53. (Previously Presented) The preservative composition of claim 52, further comprising an antimicrobial compound different from the bio-1,2-alkanediol.
54. (Previously Presented) The preservative composition of claim 42, comprising about 75% to about 99% of the bio-1,2-alkanediol.
55. (Previously Presented) The preservative composition of claim 42, wherein the bio-1,2-alkanediol comprises about 60% to about 99% bio-1,2-octanediol.

56-71. (Canceled).

72. (Currently Amended) A preservative composition comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of 7[[about 8]] to [[about]]14 carbon atoms, wherein the carbon atoms of the bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived.
73. (Currently Amended) The preservative composition of claim 72, wherein the bio-1,2-alkanediol is selected from the group consisting of: bio-1,2-heptanediol, bio-1,2-octanediol, bio-1,2-decanediol, bio-1,2-dodecanediol, and combinations thereof.
74. (Previously Presented) The preservative composition of claim 72, wherein the bio-1,2-alkanediol comprises about 60% to about 99% of bio-1,2-octanediol.
75. (Previously Presented) The preservative composition of claim 72, wherein the bio-1,2-alkanediol comprises about 75% to about 99% bio-1,2-octanediol.
76. (Previously Presented) The preservative composition of claim 72, further comprising a bio-compound selected from the group consisting of: a bio-organic acid, bio-hydroxamic acid, a bio-alkylhydroxamic acid, a bio-terpenoid compound having 5 to 20 carbon atoms, bio-1,3-propanediol, bio-1,2-butanediol, bio-1,2-pentanediol, bio-1,2-hexanediol, and combinations thereof.
77. (Previously Presented) A personal care composition comprising the preservative composition of claim 72 in a concentration of about 0.1% to about 10%.

78. (Previously Presented) The personal care composition of claim 77, wherein the preservative composition is present in a concentration of about 0.3% to about 2%.

79. (Previously Presented) The personal care composition of claim 78, wherein the preservative composition comprises about 60% to about 99% of bio-1,2-octanediol.

80. (Previously Presented) The personal care composition of claim 78, wherein the preservative composition comprises about 75% to about 99% of bio-1,2-octanediol.

81-87. (Canceled).

REMARKS

Application and Claims Status

Upon entry of the foregoing claim amendments, claims 42-55 and 72-80 are pending, and claims 1-41 and 56-71 have been cancelled.

Claims 42, 47, 72, and 73 are presently amended. No new matter has been added. While not currently under rejection, Applicants seek to further delineate claims 42 and 72 by removing the term "about" from the number of carbon atoms required by the claimed bio-1,2-alkanediol. Applicants further note that the as-filed specification at ¶ [0066] describes the use of "bio-1-heptene" as one of several starting materials for the synthesis of the claimed bio-1,2-alkanediols. Thus, the as-filed specification describes "a bio-1,2-alkanediol having a carbon chain length of 7 . . ." as required by amended claims 42 and 72, and similarly describes "bio-1,2-heptanediol" as required by claims 43 and 73.

Applicants note that the present claim amendments are made to advance the prosecution of the instant application. These amendments do not constitute any waiver or surrender of any subject matter that Applicants may be entitled to pursue, at their option, in a future submission such as a continuing or reissue application. The amendments similarly do not constitute any agreement or acquiescence as to the merits of the Office Action. Accordingly, Applicants expressly reserve the right to pursue the scope of any of the original, previously presented, withdrawn, or cancelled claims, as well as other claim scope, in a future submission.

Patentability of the Currently Amended Claims

Applicants respectfully submit that the currently amended claims are patentable over the prior art of record for the same reasons as noted in Applicants' Response to Office Action filed January 19, 2021. Specifically, the Office previously cited Wang as the most relevant prior art for the production of bio-1,2-alkanediols. Office Action dated September 17, 2020 at 3. However, as discussed in the previously submitted Response to Office Action,

> A person of ordinary skill in the art following Wang would have no way to make any 1,2-alkanediols having a chain length longer than six carbon atoms because Wang's disclosure is limited to making 1,2-alkanediols based on ring-opening hydrogenolysis of cellulose. Wang, Abstract. Nor does Wang (or any other reference) teach or suggest ways that a person of ordinary skill in the art could have modified Wang's process to create longer-chain 1,2-alkanediols. As

> such, the presently claimed compositions would not have been obvious to a person of ordinary skill in the art over Lutz in view of Wang. For these reasons, Applicants respectfully request reconsideration and withdrawal of the rejection over Lutz in view of Wang.

Response to Office Action dated January 29, 2021 at 7. In view of the fact that currently amended independent claims 42 and 72 are limited to "a carbon chain of 7 to 20" and "7 to 14 carbon atoms," respectively, Applicants submit that these claims, as currently amended, are not taught or suggested by the prior art of record for the same reasons already on record.

In the event the Office believes that an additional search is required to determine the patentability of the currently amended claims, the Examiner is invited to contact the undersigned with any questions, comments, or suggestions concerning the instant application.

CONCLUSION

In view of the foregoing amendments and remarks, it is respectfully submitted that each of the pending claims is in condition for allowance.

In the event the Commissioner determines that any fees are required for any reason, the Commissioner is hereby authorized to deduct said fee from Deposit Account No.: 50-3111/Matter No.: 484799.000.

Respectfully submitted,

Date: April 21, 2021

/*Aaron S. Lukas, Ph.D., #59443/*

Aaron S. Lukas, Ph.D.
Reg. No. 59,443
COZEN O'CONNOR
1200 Nineteenth Street N.W.
Washington, D.C. 20036
Tel: (202) 912-4823
Fax: (202) 618-4829

Electronic Acknowledgement Receipt

EFS ID:	42512798
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	171052
Filer:	Aaron Scott Lukas/Valerie Dotson
Filer Authorized By:	Aaron Scott Lukas
Attorney Docket Number:	INLX.0198U2/484799
Receipt Date:	21-APR-2021
Filing Date:	30-JAN-2019
Time Stamp:	14:46:16
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1		INLX_0198U2_484799_After_Allowance_Amendment_312.pdf	212568 568102d0cfd498cc3aa008fc4fd9a0af3db949bb	yes	6

	Multipart Description/PDF files in .zip description		
	Document Description	**Start**	**End**
	Amendment after Notice of Allowance (Rule 312)	1	1
	Claims	2	4
	Applicant Arguments/Remarks Made in an Amendment	5	6
Warnings:			
Information:			
	Total Files Size (in bytes):	212568	

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	INLX.0198U2/484799	1028

171052 7590 05/13/2021

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

EXAMINER
HOLLOMAN, NANNETTE

ART UNIT	PAPER NUMBER
1612	

NOTIFICATION DATE	DELIVERY MODE
05/13/2021	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

echoi@cozen.com
nyipdocketing@cozen.com
vdotson@cozen.com

Notice of Allowability	Application No. 16/262,902	Applicant(s) Burgo et al.	
	Examiner NANNETTE HOLLOMAN	Art Unit 1612	AIA (FITF) Status Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address--

All claims being allowable, PROSECUTION ON THE MERITS IS (OR REMAINS) CLOSED in this application. If not included herewith (or previously mailed), a Notice of Allowance (PTOL-85) or other appropriate communication will be mailed in due course. **THIS NOTICE OF ALLOWABILITY IS NOT A GRANT OF PATENT RIGHTS.** This application is subject to withdrawal from issue at the initiative of the Office or upon petition by the applicant. See 37 CFR 1.313 and MPEP 1308.

1. ☑ This communication is responsive to amandement filed 04/21/2021.
   ☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____.

2. ☐ An election was made by the applicant in response to a restriction requirement set forth during the interview on _____; the restriction requirement and election have been incorporated into this action.

3. ☑ The allowed claim(s) is/are 42-55 and 72-80 . As a result of the allowed claim(s), you may be eligible to benefit from the **Patent Prosecution Highway** program at a participating intellectual property office for the corresponding application. For more information , please see **http://www.uspto.gov/patents/init_events/pph/index.jsp** or send an inquiry to **PPHfeedback@uspto.gov.**

4. ☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

   Certified copies:

   a) ☐ All b) ☐ Some *c) ☐ None of the:

   1. ☐ Certified copies of the priority documents have been received.
   2. ☐ Certified copies of the priority documents have been received in Application No. _____ .
   3. ☐ Copies of the certified copies of the priority documents have been received in this national stage application from the International Bureau (PCT Rule 17.2(a)).

   * Certified copies not received: _____ .

Applicant has THREE MONTHS FROM THE "MAILING DATE" of this communication to file a reply complying with the requirements noted below. Failure to timely comply will result in ABANDONMENT of this application.
THIS THREE-MONTH PERIOD IS NOT EXTENDABLE.

5. ☐ CORRECTED DRAWINGS (as "replacement sheets") must be submitted.
   ☐ including changes required by the attached Examiner's Amendment / Comment or in the Office action of Paper No./Mail Date _____ .

   Identifying indicia such as the application number (see 37 CFR 1.84(c)) should be written on the drawings in the front (not the back) of each sheet. Replacement sheet(s) should be labeled as such in the header according to 37 CFR 1.121(d).

6. ☐ DEPOSIT OF and/or INFORMATION about the deposit of BIOLOGICAL MATERIAL must be submitted. Note the attached Examiner's comment regarding REQUIREMENT FOR THE DEPOSIT OF BIOLOGICAL MATERIAL.

Attachment(s)

1. ☐ Notice of References Cited (PTO-892)
2. ☐ Information Disclosure Statements (PTO/SB/08), Paper No./Mail Date _____.
3. ☐ Examiner's Comment Regarding Requirement for Deposit of Biological Material _____.
4. ☐ Interview Summary (PTO-413), Paper No./Mail Date. _____.
5. ☐ Examiner's Amendment/Comment
6. ☐ Examiner's Statement of Reasons for Allowance
7. ☑ Other PTO 271.

/NANNETTE HOLLOMAN/
Primary Examiner, Art Unit 1612

Response to Rule 312 Communication	Application No.	Applicant(s)	
	16/262,902	Burgo et al.	
	Examiner	Art Unit	AIA (FITF) Status
	NANNETTE HOLLOMAN	1612	Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

1. ☑ The amendment filed on 21 April 2021 under 37 CFR 1.312 has been considered, and has been:

a) ☑ entered.

b) ☐ entered as directed to matters of form not affecting the scope of the invention.

c) ☐ disapproved because the amendment was filed after the payment of the issue fee.

Any amendment filed after the date the issue fee is paid must be accompanied by a petition under 37 CFR 1.313(c)(1) and the required fee to withdraw the application from issue.

d) ☐ disapproved. See explanation below.

e) ☐ entered in part. See explanation below.

/NANNETTE HOLLOMAN/
Primary Examiner, Art Unit 1612

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

CPC - Searched*		
Symbol	**Date**	**Examiner**
A01N	04/10/2021	NH
C07C	04/10/2021	NH
A61K	04/10/2021	NH
A61Q	04/10/2021	NH

CPC Combination Sets - Searched*		
Symbol	**Date**	**Examiner**

US Classification - Searched*			
Class	**Subclass**	**Date**	**Examiner**

* See search history printout included with this form or the SEARCH NOTES box below to determine the scope of the search.

Search Notes		
Search Notes	**Date**	**Examiner**
PALM Inventor Search	09/13/2020	NH
EAST Search	09/13/2020	NH
NPL Search	09/13/2020	NH

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

Interference Search			
US Class/CPC Symbol	**US Subclass/CPC Group**	**Date**	**Examiner**
C07C	11/18, 2527/054, 2527/167, 2527/173, 2531/02, 65/21,	04/10/2021	NH
A61K	8/345, 2300/00, 2800/31, 8/60, 2800/412, 8/11, 8/732, 8/738, 31/197, 8/44, 8/73, 2800/48, 8/25, 8/34, 8/585, 8/92, 8/9789, 9/014, 2800/30, 31/198, 36/28, 36/534, 36/67	04/10/2021	NH
A61Q	15/00, 19/00, 13/00, 11/00, 5/00, 19/007, 19/08, 19/02, 17/005, 17/04, 19/10, 5/02, 7/00	04/10/2021	NH

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

CPC						
Symbol					Type	Version
C07C	/	29	/	04	F	2013-01-01
C07C	/	31	/	20	I	2013-01-01
C07C	/	29	/	80	I	2013-01-01
B01J	/	23	/	06	I	2013-01-01
B01J	/	37	/	08	I	2013-01-01
B01J	/	21	/	04	I	2013-01-01
C11D	/	3	/	2044	I	2013-01-01
C07C	/	1	/	24	I	2013-01-01
C07C	/	29	/	48	I	2013-01-01
C07C	/	11	/	02	I	2013-01-01
C07C	/	11	/	107	I	2013-01-01
B01J	/	2523	/	27	A	2013-01-01
C07B	/	41	/	02	A	2013-01-01
C07B	/	63	/	00	A	2013-01-01
B01J	/	2523	/	31	A	2013-01-01
A61L	/	2	/	18	A	2013-01-01
A01N	/	31	/	02	A	2013-01-01
A61K	/	8	/	345	A	2013-01-01
A61Q	/	5	/	00	A	2013-01-01
A61Q	/	11	/	00	A	2013-01-01
A61Q	/	19	/	00	A	2013-01-01
C11D	/	3	/	48	A	2013-01-01
C07C	/	2521	/	04	A	2013-01-01

CPC Combination Sets								
Symbol					Type	Set	Ranking	Version
C07C	/	1	/	24	I	1	1	2013-01-01
C07C	/	11	/	02	I	1	2	2013-01-01
C07C	/	29	/	48	I	2	1	2013-01-01
C07C	/	31	/	20	I	2	2	2013-01-01

NONE		Total Claims Allowed:	
(Assistant Examiner)	(Date)	23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	08 May 2021	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	NONE

U.S. Patent and Trademark Office

Part of Paper No.: 20210505

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

CPC Combination Sets								
Symbol					Type	Set	Ranking	Version
C07C	/	29	/	80	I	3	1	2013-01-01
C07C	/	31	/	20	I	3	2	2013-01-01

NONE		Total Claims Allowed:	
(Assistant Examiner)	(Date)	23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	08 May 2021	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	NONE

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

INTERNATIONAL CLASSIFICATION					
CLAIMED					
C07C29/04	/	29	/	04	

NON-CLAIMED				
	/		/	

US ORIGINAL CLASSIFICATION	
CLASS	SUBCLASS

CROSS REFERENCES(S)						
CLASS	SUBCLASS (ONE SUBCLASS PER BLOCK)					

NONE		Total Claims Allowed:	
(Assistant Examiner)	(Date)	23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	08 May 2021	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	NONE

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

☐ Claims renumbered in the same order as presented by applicant ☐ CPA ☐ T.D. ☐ R.1.47

CLAIMS

Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original
	1		11		21		31		41	10	51		61		71
	2		12		22		32	1	42	11	52		62	15	72
	3		13		23		33	2	43	12	53		63	16	73
	4		14		24		34	3	44	13	54		64	17	74
	5		15		25		35	4	45	14	55		65	18	75
	6		16		26		36	5	46		56		66	19	76
	7		17		27		37	6	47		57		67	20	77
	8		18		28		38	7	48		58		68	21	78
	9		19		29		39	8	49		59		69	22	79
	10		20		30		40	9	50		60		70	23	80

NONE		Total Claims Allowed:	
(Assistant Examiner)	(Date)	23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	08 May 2021	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	NONE

OK TO ENTER: /N.H/ 05/08/2021

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor: **Rocco V. Burgo**	Attorney Docket No: **INLX.0198U2/484799**
Application No.: **16/262,902**	Confirmation No.: **1028**
Filing Date: **January 20, 2019**	Art Unit: **1612**
Title: ***Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same***	Examiner: **HOLLOMAN, Nannette**

VIA EFS

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

AMENDMENT AFTER NOTICE OF ALLOWANCE UNDER 37 C.F.R. § 1.312

In response to the Notice of Allowability mailed April 14, 2021, Applicant files these papers under 37 C.F.R. § 1.312 and requests reconsideration of this application in view of the following claim amendments and remarks:

A marked-up **Listing of the Claims** begin on page 2.

Remarks begin on page 5.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor: Rocco V. Burgo

Application No.: 16/262,902 Art Unit: 1612

Filing Date: January 20, 2019 Examiner: HOLLOMAN, Nannette

Confirmation No.: 1028 Docket No: INLX.0198U2/484799

Title: **Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same**

EFS Web
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

REQUEST FOR CONTINUED EXAMINATION
INFORMATION DISCLOSURE STATEMENT UNDER §1.97(b)

Pursuant to 37 CFR § 1.56 and in accordance with 37 CFR §§ 1.97-1.98, information relating to the above-identified application is hereby disclosed. Inclusion of information in this statement is not to be construed as an admission that this information is material as that term is defined in 37 CFR § 1.56(b).

Applicants have filed this Request for Continued Examination Under 37 C.F.R. § 1.114 and accompanying Information Disclosure Statement after receiving a Communication Pursuant to Rule 114(2) EPC from the European Patent Office enclosing observations by a third party concerning the prior art to claims in co-pending application before the European Patent Office, which shares a priority claim with the present patent application.

The Request for Continued Examination Fee is attached, as well as the Information Disclosure Statement Fee of $260.00 under 37 C.F.R. § 1.17(p) is authorized to be charged to Deposit Account No. 50-3111.

Please charge any deficiency or credit any overpayment to Deposit Account No. 50-3111.

Date: July 9, 2021

/Aaron S. Lukas, Reg.# 59,443/
Aaron S. Lukas, Ph.D.
Attorney for Applicant
Reg. No. 59,443
COZEN O'CONNOR
1200 19th Street, N.W.
Washington, D.C. 20036
Tel: (202) 912-4823
Fax: (202) 861-1905

PTO/SB/30EFS (02-18)
Approved for use through 11/30/2020. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

REQUEST FOR CONTINUED EXAMINATION(RCE)TRANSMITTAL
(Submitted Only via EFS-Web)

Application Number	16262902	Filing Date	2019-01-30	Docket Number (if applicable)	INLX.0198U2/484799	Art Unit	1612
First Named Inventor	BURGO, Rocco V.			Examiner Name	HOLLOMAN, Nannette		

This is a Request for Continued Examination (RCE) under 37 CFR 1.114 of the above-identified application.
Request for Continued Examination (RCE) practice under 37 CFR 1.114 does not apply to any utility or plant application filed prior to June 8, 1995, or to any design application. The Instruction Sheet for this form is located at WWW.USPTO.GOV

SUBMISSION REQUIRED UNDER 37 CFR 1.114

Note: If the RCE is proper, any previously filed unentered amendments and amendments enclosed with the RCE will be entered in the order in which they were filed unless applicant instructs otherwise. If applicant does not wish to have any previously filed unentered amendment(s) entered, applicant must request non-entry of such amendment(s).

☐ Previously submitted. If a final Office action is outstanding, any amendments filed after the final Office action may be considered as a submission even if this box is not checked.

- ☐ Consider the arguments in the Appeal Brief or Reply Brief previously filed on _____
- ☐ Other _____

☒ Enclosed

- ☐ Amendment/Reply
- ☒ Information Disclosure Statement (IDS)
- ☐ Affidavit(s)/ Declaration(s)
- ☐ Other _____

MISCELLANEOUS

☐ Suspension of action on the above-identified application is requested under 37 CFR 1.103(c) for a period of months _____ (Period of suspension shall not exceed 3 months; Fee under 37 CFR 1.17(i) required)

☐ Other _____

FEES

☒ **The RCE fee under 37 CFR 1.17(e) is required by 37 CFR 1.114 when the RCE is filed.**
The Director is hereby authorized to charge any underpayment of fees, or credit any overpayments, to Deposit Account No ____

SIGNATURE OF APPLICANT, ATTORNEY, OR AGENT REQUIRED

☒ Patent Practitioner Signature

Applicant Signature

PTO/SB/30EFS (02-18)
Approved for use through 11/30/2020. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

Signature of Registered U.S. Patent Practitioner			
Signature	/Aaron S. Lukas, Reg.# 59443/	Date (YYYY-MM-DD)	2021-07-09
Name	Aaron S. Lukas, Ph.D.	Registration Number	59443

This collection of information is required by 37 CFR 1.114. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.11 and 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450.

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

Privacy Act Statement

The Privacy Act of 1974 (P.L. 93-579) requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C. 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether the Freedom of Information Act requires disclosure of these records.

2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.

3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.

4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).

5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.

6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).

7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.

8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspections or an issued patent.

9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

PTO/SB/08a (02-18)
Approved for use through 11/30/2020. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Not for submission under 37 CFR 1.99)

Application Number	16262902
Filing Date	2019-01-20
First Named Inventor	BURGO, Rocco V.
Art Unit	1612
Examiner Name	HOLLOMAN, Nannette
Attorney Docket Number	INLX.0198U2/484799

U.S.PATENTS Remove

Examiner Initial*	Cite No	Patent Number	Kind Code[1]	Issue Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear
	1	9242226	B2	2016-01-26	WRIGHT, Michael E.	

If you wish to add additional U.S. Patent citation information please click the Add button. Add

U.S.PATENT APPLICATION PUBLICATIONS Remove

Examiner Initial*	Cite No	Publication Number	Kind Code[1]	Publication Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear
	1	20050222276	A1	2005-10-06	SCHMAUS, Gerhard, et al.	

If you wish to add additional U.S. Published Application citation information please click the Add button. Add

FOREIGN PATENT DOCUMENTS Remove

Examiner Initial*	Cite No	Foreign Document Number[3]	Country Code[2] i	Kind Code[4]	Publication Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear	T[5]
	1							

If you wish to add additional Foreign Patent Document citation information please click the Add button Add

NON-PATENT LITERATURE DOCUMENTS Remove

Examiner Initials*	Cite No	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc), date, pages(s), volume-issue number(s), publisher, city and/or country where published.	T[5]

INFORMATION DISCLOSURE STATEMENT BY APPLICANT (Not for submission under 37 CFR 1.99)		
	Application Number	16262902
	Filing Date	2019-01-20
	First Named Inventor	BURGO, Rocco V.
	Art Unit	1612
	Examiner Name	HOLLOMAN, Nannette
	Attorney Docket Number	INLX.0198U2/484799

	1	Communication Pursuant to Rule 114(3) EPC, dated May 5, 2021 from Formalities officer Monika Bauer of the European Patent Office
	2	Third party observation on European patent application number EP3758671, filed on May 5, 2021
	3	M.J. KLUG, ET AL., Degradation of Hydrocarbons by Members of the Genus Candida, Journal of Bacteriology, Oct. 1968, p. 115-1123, Vol. 96, No. 4, printed in the USA

If you wish to add additional non-patent literature document citation information please click the Add button Add

EXAMINER SIGNATURE

Examiner Signature		Date Considered	

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through a citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

[1] See Kind Codes of USPTO Patent Documents at www.USPTO.GOV or MPEP 901.04. [2] Enter office that issued the document, by the two-letter code (WIPO Standard ST.3). [3] For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. [4] Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. [5] Applicant is to place a check mark here if English language translation is attached.

INFORMATION DISCLOSURE STATEMENT BY APPLICANT (Not for submission under 37 CFR 1.99)		
	Application Number	16262902
	Filing Date	2019-01-20
	First Named Inventor	BURGO, Rocco V.
	Art Unit	1612
	Examiner Name	HOLLOMAN, Nannette
	Attorney Docket Number	INLX.0198U2/484799

CERTIFICATION STATEMENT

Please see 37 CFR 1.97 and 1.98 to make the appropriate selection(s):

☒ That each item of information contained in the information disclosure statement was first cited in any communication from a foreign patent office in a counterpart foreign application not more than three months prior to the filing of the information disclosure statement. See 37 CFR 1.97(e)(1).

OR

☐ That no item of information contained in the information disclosure statement was cited in a communication from a foreign patent office in a counterpart foreign application, and, to the knowledge of the person signing the certification after making reasonable inquiry, no item of information contained in the information disclosure statement was known to any individual designated in 37 CFR 1.56(c) more than three months prior to the filing of the information disclosure statement. See 37 CFR 1.97(e)(2).

See attached certification statement.

☒ The fee set forth in 37 CFR 1.17 (p) has been submitted herewith.

A certification statement is not submitted herewith.

SIGNATURE

A signature of the applicant or representative is required in accordance with CFR 1.33, 10.18. Please see CFR 1.4(d) for the form of the signature.

Signature	/Aaron S. Lukas, Ph.D. - 59,443/	Date (YYYY-MM-DD)	2021-07-09
Name/Print	Aaron S. Lukas, Ph.D.	Registration Number	59,443

This collection of information is required by 37 CFR 1.97 and 1.98. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 1 hour to complete, including gathering, preparing and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

Privacy Act Statement

The Privacy Act of 1974 (P.L. 93-579) requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C. 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether the Freedom of Information Act requires disclosure of these record s.

2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.

3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.

4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).

5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.

6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).

7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.

8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspections or an issued patent.

9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor: Rocco V. Burgo

Application No.: 16/262,902 Art Unit: 1612

Filing Date: January 20, 2019 Examiner: HOLLOMAN, Nannette

Confirmation No.: 1028 Docket No: INLX.0198U2/484799

Title: **Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same**

EFS Web
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

REQUEST FOR CONTINUED EXAMINATION
INFORMATION DISCLOSURE STATEMENT UNDER §1.97(b)

Pursuant to 37 CFR § 1.56 and in accordance with 37 CFR §§ 1.97-1.98, information relating to the above-identified application is hereby disclosed. Inclusion of information in this statement is not to be construed as an admission that this information is material as that term is defined in 37 CFR § 1.56(b).

Applicants have filed this Request for Continued Examination Under 37 C.F.R. § 1.114 and accompanying Information Disclosure Statement after receiving a Communication Pursuant to Rule 114(2) EPC from the European Patent Office enclosing observations by a third party concerning the prior art to claims in co-pending application before the European Patent Office, which shares a priority claim with the present patent application.

The Request for Continued Examination Fee is attached, as well as the Information Disclosure Statement Fee of $260.00 under 37 C.F.R. § 1.17(p) is authorized to be charged to Deposit Account No. 50-3111.

Please charge any deficiency or credit any overpayment to Deposit Account No. 50-3111.

Date: July 9, 2021

/Aaron S. Lukas, Reg.# 59,443/
Aaron S. Lukas, Ph.D.
Attorney for Applicant
Reg. No. 59,443
COZEN O'CONNOR
1200 19th Street, N.W.
Washington, D.C. 20036
Tel: (202) 912-4823
Fax: (202) 861-1905

Electronic Patent Application Fee Transmittal

Application Number:	16262902
Filing Date:	30-Jan-2019
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Filer:	Aaron Scott Lukas/Valerie Dotson
Attorney Docket Number:	INLX.0198U2/484799

Filed as Large Entity

Filing Fees for Utility under 35 USC 111(a)

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Pages:				
Claims:				
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
Extension-of-Time:				

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Miscellaneous:				
RCE- 1ST REQUEST	1801	1	1360	1360
SUBMISSION- INFORMATION DISCLOSURE STMT	1806	1	260	260
		Total in USD ($)		**1620**

Electronic Acknowledgement Receipt

EFS ID:	43206811
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	171052
Filer:	Aaron Scott Lukas/Valerie Dotson
Filer Authorized By:	Aaron Scott Lukas
Attorney Docket Number:	INLX.0198U2/484799
Receipt Date:	09-JUL-2021
Filing Date:	30-JAN-2019
Time Stamp:	10:23:00
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	CARD
Payment was successfully received in RAM	$1620
RAM confirmation Number	E202179A23482990
Deposit Account	503111
Authorized User	Valerie Dotson

The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows:

37 CFR 1.21 (Miscellaneous fees and charges)

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Request for Continued Examination (RCE)	INLX_0198U2_484799_Request_for_Continued_Examination.pdf	1350000 f07ed3182df1c0f51a4a3822b02681f81550698e	no	3
Warnings:					
Information:					
2	Information Disclosure Statement (IDS) Form (SB08)	INLX_0198U2_484799_Information_Disclosure_Statement.pdf	1042074 43e08efc5423b75d4ec6dc478c178ebedd82a7af	no	4
Warnings:					
Information:					
3	Non Patent Literature	INLX_0198U2_484799_Communication_Pursuant_to_Rule_114.PDF	596127 9e95db2d93d55ebb2796e547d820a0f0f571f2fa	no	10
Warnings:					
Information:					
4	Non Patent Literature	INLX_0198U2_484799_Third_Party_Observation_European_Patent_Application.PDF	688043 aa292b1cae4634995f7cf3ea4cee0f8a56a8fed6	no	14
Warnings:					
Information:					
5	Non Patent Literature	INLX_0198U2_484799_NPL_Klug_1968.pdf	691079 9b654e766b63b7319c81c14f7843d19146ceaa03	no	9
Warnings:					
Information:					
6	Transmittal Letter	INLX_0198U2_484799_Transmittal.pdf	144040 04855cd260bde1e655b9bcf5e327598dda6aa0ac	no	2
Warnings:					
Information:					

7	Fee Worksheet (SB06)	fee-info.pdf	32437 b6c096dd1b65af6b4cfc13f798b98eadae073c1c	no	2
Warnings:					
Information:					
			Total Files Size (in bytes):	4543800	

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	INLX.0198U2/484799	1028

171052 7590 08/31/2021

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

EXAMINER
HOLLOMAN, NANNETTE

ART UNIT	PAPER NUMBER
1612	

NOTIFICATION DATE	DELIVERY MODE
08/31/2021	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

echoi@cozen.com
nyipdocketing@cozen.com
vdotson@cozen.com

Office Action Summary	Application No. 16/262,902	Applicant(s) Burgo et al.	
	Examiner NANNETTE HOLLOMAN	Art Unit 1612	AIA (FITF) Status Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTHS FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

1)☑ Responsive to communication(s) filed on 07092021.

☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____.

2a)☐ This action is **FINAL.** 2b) ☑ This action is non-final.

3)☐ An election was made by the applicant in response to a restriction requirement set forth during the interview on _____; the restriction requirement and election have been incorporated into this action.

4)☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims*

5) ☑ Claim(s) 42-55 and 72-80 is/are pending in the application.

5a) Of the above claim(s) _____ is/are withdrawn from consideration.

6) ☐ Claim(s) _____ is/are allowed.

7) ☑ Claim(s) 42-55 and 72-80 is/are rejected.

8) ☐ Claim(s) _____ is/are objected to.

9) ☐ Claim(s) _____ are subject to restriction and/or election requirement

* If any claims have been determined allowable, you may be eligible to benefit from the **Patent Prosecution Highway** program at a participating intellectual property office for the corresponding application. For more information, please see http://www.uspto.gov/patents/init_events/pph/index.jsp or send an inquiry to **PPHfeedback@uspto.gov.**

Application Papers

10)☐ The specification is objected to by the Examiner.

11)☐ The drawing(s) filed on _____ is/are: a)☐ accepted or b)☐ objected to by the Examiner.

Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).

Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).

Priority under 35 U.S.C. § 119

12)☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

Certified copies:

a)☐ All b)☐ Some** c)☐ None of the:

1.☐ Certified copies of the priority documents have been received.

2.☐ Certified copies of the priority documents have been received in Application No. _____.

3.☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

** See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

1) ☐ Notice of References Cited (PTO-892)

2) ☑ Information Disclosure Statement(s) (PTO/SB/08a and/or PTO/SB/08b) Paper No(s)/Mail Date _____.

3) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date _____.

4) ☐ Other: _____.

DETAILED ACTION

Notice of Pre-AIA or AIA Status

The present application, filed on or after March 16, 2013, is being examined under the first inventor to file provisions of the AIA.

This Office Action is in response to the Request for Continued Examination filed on 07/09/2021. Applicants' arguments, filed 07/09/2021, have been fully considered. Rejections and/or objections not reiterated from previous office actions are hereby withdrawn. The following rejections and/or objections are either reiterated or newly applied. They constitute the complete set presently being applied to the instant application.

The text of those sections of Title 35, U.S. Code not included in this action can be found in a prior Office Action.

Claim Rejections - 35 USC § 103

In the event the determination of the status of the application as subject to AIA 35 U.S.C. 102 and 103 (or as subject to pre-AIA 35 U.S.C. 102 and 103) is incorrect, any correction of the statutory basis for the rejection will not be considered a new ground of rejection if the prior art relied upon, and the rationale supporting the rejection, would be the same under either status.

The following is a quotation of 35 U.S.C. 103 which forms the basis for all obviousness rejections set forth in this Office action:

> A patent for a claimed invention may not be obtained, notwithstanding that the claimed invention is not identically disclosed as set forth in section 102, if the differences between the claimed invention and the prior art are such that the claimed invention as a whole would have been obvious before the effective filing date of the claimed invention to a person having ordinary skill in the art to which the claimed invention pertains. Patentability shall not be negated by the manner in which the invention was made.

The factual inquiries for establishing a background for determining obviousness under 35 U.S.C. 103 are summarized as follows:

1. Determining the scope and contents of the prior art.

2. Ascertaining the differences between the prior art and the claims at issue.

3. Resolving the level of ordinary skill in the pertinent art.

4. Considering objective evidence present in the application indicating obviousness or nonobviousness.

This application currently names joint inventors. In considering patentability of the claims the examiner presumes that the subject matter of the various claims was commonly owned as of the effective filing date of the claimed invention(s) absent any evidence to the contrary. Applicant is advised of the obligation under 37 CFR 1.56 to point out the inventor and effective filing dates of each claim that was not commonly owned as of the effective filing date of the later invention in order for the examiner to consider the applicability of 35 U.S.C. 102(b)(2)(C) for any potential 35 U.S.C. 102(a)(2) prior art against the later invention.

Claims 42-44, 47-55 and 72-80 is/are rejected under 35 U.S.C. 103 as being unpatentable over Schmaus et al. (US Patent Pub. 2005/0222276) in view of Wang et al. (Green Chem., 2012, 14, pp. 758-765, previously disclosed).

Schmaus et al. disclose the use of mixtures of two, three or more 1,2-alkanediols with chain links of 5 to 10 as antimicrobial active compounds (ABSTRACT). Schmaus et al. disclose the use of a mixture of 1,2-hexanediol and 1,2-octandediol; herein the proportion of each individual diol should be in the range of 1 to 99% (m/m) ([0032] and [0033]). Schmaus et al. disclose the mixtures ate used for the preservation of perishable products such, as cosmetics, pharmaceutical products and foods ([0034]).

Schmaus et al. differs from the instant claims insofar as they do not disclose wherein the 1,2-alkanediols are bio-1,2-alkanediols.

Wang et al. disclose due to the diminishing fossil fuel reservoirs together with the ever deteriorating environment associated with the worldwide fossil fuel based industry, it is imperative to make use of alternative feedstock for production of chemicals and fuels (p.758, Introduction).

It would have been obvious to one of ordinary skill in the art before the effective filing date of the claimed invention to have used "bio" based 1,2-alkanediols in the formulation of Schmaus motivated by the desire to preserve the environment as taught by Wang.

Claim 42-55 and 72-80 is/are rejected under 35 U.S.C. 103 as being unpatentable over Lutz (US Patent Pub. 2012/0282207, previously disclosed) in view of Wang et al. (Green Chem., 2012, 14, pp 758-765, previously disclosed).

Lutz discloses an antimicrobial compositions and a product comprising a preservative effective amount of the said composition (Abstract). Lutz discloses household; fabric care and pharmaceutical products ([0043] and claim 25). Lutz discloses the composition comprises caprylyl glycol, which meets the limitation of 1,2-octanediol, pentylene glycol, hexylene glycol, and any combination thereof (reference claim 7). Lutz discloses the composition further comprises an antimicrobial mixture (reference claim 1).

Lutz differs from the instant claims insofar as it does not disclose the 1,2-alkanediol as "bio".

Wang et al. disclose due to the diminishing fossil fuel reservoirs together with the ever-deteriorating environment associated with the worldwide fossil-fuel-based industry, it is imperative to make use of alternative feedstock for the production of chemicals and fuels (p. 758, Introduction).

It would have been obvious to one of ordinary skill in the art before the effective filing date of the claimed invention to have used "bio" based 1,2-alkanediols in the formulation of Lutz motivated by the desire to preserve the environment as taught by Wang.

Conclusion

No claims are allowed.

Any inquiry concerning this communication or earlier communications from the examiner should be directed to NANNETTE HOLLOMAN whose telephone number is (571)270-5231. The examiner can normally be reached on Monday-Friday 9am-6pm.

Examiner interviews are available via telephone, in-person, and video conferencing using a USPTO supplied web-based collaboration tool. To schedule an interview, applicant is encouraged to use the USPTO Automated Interview Request (AIR) at http://www.uspto.gov/interviewpractice.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Frederick Krass can be reached on (571)272-0580. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see https://ppair-my.uspto.gov/pair/PrivatePair. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

/NANNETTE HOLLOMAN/
Primary Examiner, Art Unit 1612

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

CPC - Searched*		
Symbol	**Date**	**Examiner**
A01N	04/10/2021	NH
C07C	04/10/2021	NH
A61K	04/10/2021	NH
A61Q	04/10/2021	NH

CPC Combination Sets - Searched*		
Symbol	**Date**	**Examiner**

US Classification - Searched*			
Class	**Subclass**	**Date**	**Examiner**

* See search history printout included with this form or the SEARCH NOTES box below to determine the scope of the search.

Search Notes		
Search Notes	**Date**	**Examiner**
PALM Inventor Search	09/13/2020	NH
EAST Search	09/13/2020	NH
NPL Search	09/13/2020	NH
PALM Inventor Search	08/26/2021	NH
EAST Search	08/26/2021	NH
NPL Search	08/26/2021	NH

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

Interference Search			
US Class/CPC Symbol	**US Subclass/CPC Group**	**Date**	**Examiner**
C07C	11/18, 2527/054, 2527/167, 2527/173, 2531/02, 65/21,	04/10/2021	NH
A61K	8/345, 2300/00, 2800/31, 8/60, 2800/412, 8/11, 8/732, 8/738, 31/197, 8/44, 8/73, 2800/48, 8/25, 8/34, 8/585, 8/92, 8/9789, 9/014, 2800/30, 31/198, 36/28, 36/534, 36/67	04/10/2021	NH
A61Q	15/00, 19/00, 13/00, 11/00, 5/00, 19/007, 19/08, 19/02, 17/005, 17/04, 19/10, 5/02, 7/00	04/10/2021	NH

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

EAST Search History

EAST Search History (Prior Art)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
S1	937	((("BURGO") near3 ("Rocco")) OR (("WRIGHT") near3 ("Michael")) OR (("MOSSER") near3 ("Gary")) OR (("FEVOLA") near3 ("Michael"))).INV.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	OFF	2020/07/07 19:12
S2	378	(bio with alkene) bioalkene	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/07/07 19:19
S3	6,577	regioselectivity	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/09 14:30
S4	13	regioselectivity same alkanediol	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/09 14:30
S5	7	regioselectivity same (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/09 14:47
S6	1	("7385092").PN.	USPAT; USOCR	OR	OFF	2020/09/09 14:53
S7	2	wo-2016007196-$.did.	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	OFF	2020/09/11 11:19
S8	6	alkanediol same (household and pharmaceutical and cosmetic)	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:25
S9	3	(household and pharmaceutical and cosmetic) same (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26

S10	336	(household and pharmaceutical and cosmetic) and (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26
S11	6	((household and pharmaceutical and cosmetic) and (octanediol or (caprylyl adj glycol))).clm.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26
S12	11	((household and pharmaceutical and cosmetic) and (octanediol or ((caprylyl or hexylene or decylene) adj glycol) or alkanediol)).clm.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:47
S13	946	((("BURGO") near3 ("Rocco")) OR (("WRIGHT") near3 ("Michael")) OR (("MOSSER") near3 ("Gary")) OR (("FEVOLA") near3 ("Michael"))).INV.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	OFF	2020/09/11 19:14
S14	3	S13 and alkanediol	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/09/11 19:18
S15	7	(("8912373") or ("9856461") or ("4234752") or ("20050065389") or ("20030065233") or ("20060161025") or ("20140066666")).PN.	US-PGPUB; USPAT; USOCR	OR	OFF	2020/09/13 13:37
S16	42,261	decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/02/27 23:31
S17	13,209	S16 and (preservative or pharmaceutical or cosmetic)	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/02/27 23:32
S18	1,488	S17 and "bio"	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/02/27 23:32
S19	67	"bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol)))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/04/10 19:56

S20	549,665	((A01N47/44 OR A01N37/36 OR A01N59/16 OR A01N31/02 OR A01N25/34 OR A01N59/00 OR A01N61/00 OR A01N25/00 OR A01N2300/00 OR A01N25/10 OR C07C31/20 OR C07C1/24 OR C07C11/02 OR C07C29/48 OR C07C29/80 OR C07C263/12 OR C07C265/14 OR C07C2521/04 OR C07C11/107 OR C07C29/04 OR C07C247/24 OR C07C11/167 OR C07C11/18 OR C07C2527/054 OR C07C2527/167 OR C07C2527/173 OR C07C2531/02 OR C07C65/21 OR A61K8/345 OR A61K2300/00 OR A61K2800/31 OR A61K8/60 OR A61K2800/412 OR A61K8/11 OR A61K8/732 OR A61K8/738 OR A61K31/197 OR A61K8/44 OR A61K8/73 OR A61K2800/48 OR A61K8/25 OR A61K8/34 OR A61K8/585 OR A61K8/92 OR A61K8/9789 OR A61K9/0014 OR A61K2800/30 OR A61K31/198 OR A61K36/28 OR A61K36/534 OR A61K36/67 OR A61K36/886 OR A61K45/06 OR A61K47/10 OR A61K8/24 OR A61K8/362 OR A61K8/37 OR A61K8/41 OR A61K8/42 OR A61K8/4973 OR A61K8/498 OR A61K8/891 OR A61K9/06 OR A61K2800/10 OR A61K2800/28 OR A61K2800/524 OR A61K2800/56 OR A61K2800/87 OR A61K2800/874 OR A61K31/327 OR A61K47/22 OR A61K47/44 OR A61K47/46 OR A61K8/0216 OR A61K8/046 OR A61K8/19 OR A61K8/20 OR A61K8/23 OR A61K8/26 OR A61K8/31 OR A61K8/64 OR A61K8/678 OR A61K8/735 OR A61K8/8111 OR A61K8/8147 OR A61K8/893 OR A61K8/895 OR A61K8/922 OR A61K8/9794 OR A61K9/107 OR A61K2035/115 OR A61K2800/43 OR A61K2800/522 OR A61K2800/592 OR A61K2800/5922 OR A61K2800/594 OR A61K2800/596 OR A61K2800/624 OR A61K2800/805 OR A61K31/192 OR A61K31/355 OR A61K31/60 OR A61K33/30 OR A61K35/745 OR A61K35/747 OR A61K36/185 OR A61K36/53 OR A61K47/02 OR A61K47/06 OR A61K47/12 OR A61K47/14 OR A61K47/183 OR A61K47/24 OR	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/04/10 19:57

		A61K47/34 OR A61K47/38 OR A61K49/0006 OR A61K49/0008 OR A61K8/06 OR A61K8/062 OR A61K8/28 OR A61K8/29 OR A61K8/342 OR A61K8/347 OR A61K8/365 OR A61K8/368 OR A61K8/375 OR A61K8/416 OR A61K8/445 OR A61K8/463 OR A61K8/49 OR A61K8/4926 OR A61K8/63 OR A61K8/66 OR A61K8/675 OR A61K8/8152 OR A61K8/8194 OR A61K8/927 OR A61K8/965 OR A61K8/97 OR A61K9/08 OR A61Q15/00 OR A61Q19/00 OR A61Q13/00 OR A61Q11/00 OR A61Q5/00 OR A61Q19/007 OR A61Q19/08 OR A61Q19/02 OR A61Q17/005 OR A61Q17/04 OR A61Q19/10 OR A61Q5/02 OR A61Q7/00).CPC.)				
S21	42	S20 and ("bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/04/10 19:57
S24	2	(("9242226") or ("20050222276")).PN.	US-PGPUB; USPAT; USOCR	OR	OFF	2021/08/14 21:51

EAST Search History (Interference)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
S22	125,400	((A01N47/44 OR A01N37/36 OR A01N59/16 OR A01N31/02 OR A01N25/34 OR A01N59/00 OR A01N61/00 OR A01N25/00 OR A01N2300/00 OR A01N25/10 OR C07C31/20 OR C07C1/24 OR C07C11/02 OR C07C29/48 OR C07C29/80 OR C07C263/12 OR C07C265/14 OR C07C2521/04 OR C07C11/107 OR C07C29/04 OR C07C247/24 OR C07C11/167 OR C07C11/18 OR C07C2527/054 OR C07C2527/167 OR C07C2527/173 OR C07C2531/02 OR C07C65/21 OR A61K8/345 OR A61K2300/00 OR A61K2800/31 OR A61K8/60 OR A61K2800/412 OR A61K8/11 OR A61K8/732 OR A61K8/738 OR A61K31/197 OR A61K8/44 OR A61K8/73 OR A61K2800/48 OR A61K8/25 OR A61K8/34 OR A61K8/585 OR A61K8/92 OR A61K8/9789 OR A61K9/0014 OR A61K2800/30 OR A61K31/198 OR A61K36/28	USPAT	OR	ON	2021/04/10 19:59

		OR A61K36/534 OR A61K36/67 OR A61K36/886 OR A61K45/06 OR A61K47/10 OR A61K8/24 OR A61K8/362 OR A61K8/37 OR A61K8/41 OR A61K8/42 OR A61K8/4973 OR A61K8/498 OR A61K8/891 OR A61K9/06 OR A61K2800/10 OR A61K2800/28 OR A61K2800/524 OR A61K2800/56 OR A61K2800/87 OR A61K2800/874 OR A61K31/327 OR A61K47/22 OR A61K47/44 OR A61K47/46 OR A61K8/0216 OR A61K8/046 OR A61K8/19 OR A61K8/20 OR A61K8/23 OR A61K8/26 OR A61K8/31 OR A61K8/64 OR A61K8/678 OR A61K8/735 OR A61K8/8111 OR A61K8/8147 OR A61K8/893 OR A61K8/895 OR A61K8/922 OR A61K8/9794 OR A61K9/107 OR A61K2035/115 OR A61K2800/43 OR A61K2800/522 OR A61K2800/592 OR A61K2800/5922 OR A61K2800/594 OR A61K2800/596 OR A61K2800/624 OR A61K2800/805 OR A61K31/192 OR A61K31/355 OR A61K31/60 OR A61K33/30 OR A61K35/745 OR A61K35/747 OR A61K36/185 OR A61K36/53 OR A61K47/02 OR A61K47/06 OR A61K47/12 OR A61K47/14 OR A61K47/183 OR A61K47/24 OR A61K47/34 OR A61K47/38 OR A61K49/0006 OR A61K49/0008 OR A61K8/06 OR A61K8/062 OR A61K8/28 OR A61K8/29 OR A61K8/342 OR A61K8/347 OR A61K8/365 OR A61K8/368 OR A61K8/375 OR A61K8/416 OR A61K8/445 OR A61K8/463 OR A61K8/49 OR A61K8/4926 OR A61K8/63 OR A61K8/66 OR A61K8/675 OR A61K8/8152 OR A61K8/8194 OR A61K8/927 OR A61K8/965 OR A61K8/97 OR A61K9/08 OR A61Q15/00 OR A61Q19/00 OR A61Q13/00 OR A61Q11/00 OR A61Q5/00 OR A61Q19/007 OR A61Q19/08 OR A61Q19/02 OR A61Q17/005 OR A61Q17/04 OR A61Q19/10 OR A61Q5/02 OR A61Q7/00).CPC.)				
S23	10	S22 and ("bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))))	USPAT	OR	ON	2021/04/10 19:59

8/26/2021 8:02:36 PM
C:\Users\nholloman\Documents\EAST\Workspaces\16262902.wsp

Doc code: IDS
Doc description: Information Disclosure Statement (IDS) Filed

PTO/SB/08a (02-18)
Approved for use through 11/30/2020. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Not for submission under 37 CFR 1.99)

Application Number	16262902
Filing Date	2019-01-20
First Named Inventor	BURGO, Rocco V.
Art Unit	1612
Examiner Name	HOLLOMAN, Nannette
Attorney Docket Number	INLX.0198U2/484799

U.S.PATENTS Remove

Examiner Initial*	Cite No	Patent Number	Kind Code[1]	Issue Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear
	1	9242226	B2	2016-01-26	WRIGHT, Michael E.	

If you wish to add additional U.S. Patent citation information please click the Add button. Add

U.S.PATENT APPLICATION PUBLICATIONS Remove

Examiner Initial*	Cite No	Publication Number	Kind Code[1]	Publication Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear
	1	20050222276	A1	2005-10-06	SCHMAUS, Gerhard, et al.	

If you wish to add additional U.S. Published Application citation information please click the Add button. Add

FOREIGN PATENT DOCUMENTS Remove

Examiner Initial*	Cite No	Foreign Document Number[3]	Country Code[2] i	Kind Code[4]	Publication Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear	T[5]
	1							

If you wish to add additional Foreign Patent Document citation information please click the Add button Add

NON-PATENT LITERATURE DOCUMENTS Remove

Examiner Initials*	Cite No	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc), date, pages(s), volume-issue number(s), publisher, city and/or country where published.	T[5]

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Not for submission under 37 CFR 1.99)

Application Number	16262902
Filing Date	2019-01-20
First Named Inventor	BURGO, Rocco V.
Art Unit	1612
Examiner Name	HOLLOMAN, Nannette
Attorney Docket Number	INLX.0198U2/484799

	1	Communication Pursuant to Rule 114(3) EPC, dated May 5, 2021 from Formalities officer Monika Bauer of the European Patent Office
	2	Third party observation on European patent application number EP3758671, filed on May 5, 2021
	3	M.J. KLUG, ET AL., Degradation of Hydrocarbons by Members of the Genus Candida, Journal of Bacteriology, Oct. 1968, p. 115-1123, Vol. 96, No. 4, printed in the USA

If you wish to add additional non-patent literature document citation information please click the Add button Add

EXAMINER SIGNATURE

Examiner Signature	/NANNETTE HOLLOMAN/	Date Considered	08/14/2021

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through a citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

[1] See Kind Codes of USPTO Patent Documents at www.USPTO.GOV or MPEP 901.04. [2] Enter office that issued the document, by the two-letter code (WIPO Standard ST.3). [3] For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. [4] Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. [5] Applicant is to place a check mark here if English language translation is attached.

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Not for submission under 37 CFR 1.99)

Application Number	16262902
Filing Date	2019-01-20
First Named Inventor	BURGO, Rocco V.
Art Unit	1612
Examiner Name	HOLLOMAN, Nannette
Attorney Docket Number	INLX.0198U2/484799

CERTIFICATION STATEMENT

Please see 37 CFR 1.97 and 1.98 to make the appropriate selection(s):

X That each item of information contained in the information disclosure statement was first cited in any communication from a foreign patent office in a counterpart foreign application not more than three months prior to the filing of the information disclosure statement. See 37 CFR 1.97(e)(1).

OR

☐ That no item of information contained in the information disclosure statement was cited in a communication from a foreign patent office in a counterpart foreign application, and, to the knowledge of the person signing the certification after making reasonable inquiry, no item of information contained in the information disclosure statement was known to any individual designated in 37 CFR 1.56(c) more than three months prior to the filing of the information disclosure statement. See 37 CFR 1.97(e)(2).

See attached certification statement.

X The fee set forth in 37 CFR 1.17 (p) has been submitted herewith.

A certification statement is not submitted herewith.

SIGNATURE

A signature of the applicant or representative is required in accordance with CFR 1.33, 10.18. Please see CFR 1.4(d) for the form of the signature.

Signature	/Aaron S. Lukas, Ph.D. - 59,443/	Date (YYYY-MM-DD)	2021-07-09
Name/Print	Aaron S. Lukas, Ph.D.	Registration Number	59,443

This collection of information is required by 37 CFR 1.97 and 1.98. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 1 hour to complete, including gathering, preparing and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

Privacy Act Statement

The Privacy Act of 1974 (P.L. 93-579) requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C. 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether the Freedom of Information Act requires disclosure of these record s.

2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.

3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.

4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).

5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.

6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).

7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.

8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspections or an issued patent.

9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor: **Rocco V. Burgo**	Attorney Docket No: **INLX.0198U2/484799**
Application No.: **16/262,902**	Confirmation No.: **1028**
Filing Date: **January 20, 2019**	Art Unit: **1612**
Title: ***Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same***	Examiner: **HOLLOMAN, Nannette**

VIA EFS

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

RESPONSE TO OFFICE ACTION UNDER 37 C.F.R. § 1.111

In response to the Non-Final Office Action mailed August 31, 2021, Applicants request reconsideration of this application in view of the following claim amendments and remarks:

A marked-up **Listing of the Claims** begin on page 2.

Remarks begin on page 5.

LISTING OF THE CLAIMS

This listing of claims will replace all prior versions, and listings, of claims in the application.

What is Claimed is:

1-41. (Canceled)

42. (Previously Presented) A preservative composition, comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of 7 to 20 carbon atoms, wherein the carbon atoms of the first bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived.

43. (Previously Presented) A personal care composition comprising the preservative composition of claim 42.

44. (Previously Presented) The personal care composition of claim 43, wherein the personal care composition is a hair care composition, an oral care composition, a skin care composition or a cosmetic composition.

45. (Previously Presented) The personal care composition of claim 43, wherein the personal care composition is a household product.

46. (Previously Presented) The personal care composition of claim 45, wherein the household product is a fabric care product or a cleaning product.

47. (Previously Presented) The personal care composition of claim 43, wherein the bio-1,2-alkanediol is selected from the group consisting of: bio-1,2-heptanediol, bio-1,2-octanediol, bio-1,2-decanediol, bio-1,2-dodecanediol, and combinations thereof.

48. (Previously Presented) The personal care composition of claim 43, wherein the personal care composition is a pharmaceutical composition, a vitamin composition, or a health care composition.

49. (Previously Presented) The personal care composition of claim 43, wherein the preservative composition is present in a concentration of about 0.1% to about 10%.

50. (Previously Presented) The personal care composition of claim 43, further comprising a bio-compound selected from the group consisting of: a bio-organic acid, bio-hydroxamic acid, a bio-alkylhydroxamic acid, a bio-terpenoid compound having 5 to 20 carbon atoms, bio-1,3-propanediol, bio-1,2-butanediol, bio-1,2-pentanediol, and combinations thereof.

51. (Previously Presented) The personal care composition of claim 50, wherein the preservative composition is present in a concentration of about 0.1% to about 10%.
52. (Previously Presented) The preservative composition of claim 42, wherein the composition further comprises at least one other bio-compound different from the bio-1,2-alkanediol.
53. (Previously Presented) The preservative composition of claim 52, further comprising an antimicrobial compound different from the bio-1,2-alkanediol.
54. (Previously Presented) The preservative composition of claim 42, comprising about 75% to about 99% of the bio-1,2-alkanediol.
55. (Previously Presented) The preservative composition of claim 42, wherein the bio-1,2-alkanediol comprises about 60% to about 99% bio-1,2-octanediol.

56-71. (Canceled).

72. (Previously Presented) A preservative composition comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of 7 to 14 carbon atoms, wherein the carbon atoms of the bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived.
73. (Previously Presented) The preservative composition of claim 72, wherein the bio-1,2-alkanediol is selected from the group consisting of: bio-1,2-heptanediol, bio-1,2-octanediol, bio-1,2-decanediol, bio-1,2-dodecanediol, and combinations thereof.
74. (Previously Presented) The preservative composition of claim 72, wherein the bio-1,2-alkanediol comprises about 60% to about 99% of bio-1,2-octanediol.
75. (Previously Presented) The preservative composition of claim 72, wherein the bio-1,2-alkanediol comprises about 75% to about 99% bio-1,2-octanediol.
76. (Previously Presented) The preservative composition of claim 72, further comprising a bio-compound selected from the group consisting of: a bio-organic acid, bio-hydroxamic acid, a bio-alkylhydroxamic acid, a bio-terpenoid compound having 5 to 20 carbon atoms, bio-1,3-propanediol, bio-1,2-butanediol, bio-1,2-pentanediol, bio-1,2-hexanediol, and combinations thereof.
77. (Previously Presented) A personal care composition comprising the preservative composition of claim 72 in a concentration of about 0.1% to about 10%.

78. (Previously Presented) The personal care composition of claim 77, wherein the preservative composition is present in a concentration of about 0.3% to about 2%.

79. (Previously Presented) The personal care composition of claim 78, wherein the preservative composition comprises about 60% to about 99% of bio-1,2-octanediol.

80. (Previously Presented) The personal care composition of claim 78, wherein the preservative composition comprises about 75% to about 99% of bio-1,2-octanediol.

81-87. (Canceled).

REMARKS

Application and Claims Status

Upon entry of the foregoing claim amendments, claims 42-55 and 72-80 are pending, and claims 1-41 and 56-71 were previously cancelled.

Claims 42-55 and 72-80 are rejected. In view of the following remarks, Applicants respectfully submit that the pending rejections have been traversed and respectfully request favorable reconsideration and allowance of the pending claims.

Patentability of the Claims

35 U.S.C. § 103, Obviousness

The Office has rejected claims 42-44, 47-55, and 72-80 under 35 U.S.C. § 103 as allegedly obvious over Schmaus (US Patent Pub. No. 2005/0222276) in view of Wang (*Green Chem.*, 14:758-65 (2012). Office Action at 2-4.

The Office has also rejected claims 42-55 and 72-80 under 35 U.S.C. § 103 as allegedly obvious over Lutz (US Patent Pub. No. 2012/0282207) in view of Wang. Office Action at 4-5.

Schmaus discloses antimicrobial composition that are mixtures of two, three, or more straight-chain 1,2-alkanediols having chain lengths of 5 to 10 carbon atoms. *See* Schmaus, at e.g., Abstract, ¶¶ [0012]-[0017]. The Office acknowledges that Schmaus does not disclose the use of "bio" alcohols such as the presently claimed bio-1,2-alkanediols. Office Action at 3-4. The Office asserts that a person ordinary skill in the art would have relied on the disclosure of Wang to prepare bio-1,2-alkanediols for use in the compositions of Schmaus, which impliedly, a person of ordinary skill in the art would have used to prepare the claimed compositions. Office Action at 4.

Lutz discloses antimicrobial compositions that contain two or more "natural or naturally derived" fatty acids that can be mixed with other "natural or naturally derived products," an alcoholic solvent, and/or an organic acid. *See* Lutz, Abstract, ¶¶ [0006]-[0012]. The Office acknowledges that Lutz does not disclose the use of "bio" alcohols such as the presently claimed bio-1,2-alkanediols. Office Action at 3. The Office asserts that a person ordinary skill in the art would have relied on the disclosure of Wang to prepare bio-1,2-alkanediols for use in the compositions of Lutz, which impliedly, a person of ordinary skill in the art would have used to prepare the claimed compositions. Office Action at 4-5.

Applicants respectfully traverse these rejections.

The present independent claims 42 and 72 require "[a] preservative composition comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of 7 to 20 carbon atoms, wherein the carbon atoms of the bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived." Schmaus and Lutz admittedly fail to teach or suggest how to prepare any bio-1,2-alkanediols, let alone bio-1,2-alkanediols having a chain length of 7 to 20 carbon atoms. The Office relies solely on Wang as allegedly teaching this aspect of the claimed invention. However, Wang is limited in that it only discloses methods to of using cellulosic feedstocks to prepare compounds that include bio-C_2-C_6 alkanediols,. *See* Wang at 761, Table 1. That is, Wang's processes are limited to the production of 1,2-alkanediols "with carbon number[s] in the range of 2-6." Wang at 759, col. 1. Conversely, the current claims require bio-1,2-alkanediols having a carbon chain length of 7 to 20 carbon atoms. A person of ordinary skill in the art following Wang would have no way to make any 1,2-alkanediols having a chain length longer than six carbon atoms because Wang's disclosure is limited to making 1,2-alkanediols based on ring-opening hydrogenolysis of cellulose. Wang, Abstract. Nor does Wang (or any other reference) teach or suggest ways that a person of ordinary skill in the art could have modified Wang's process to create longer-chain 1,2-alkanediols. As such, the presently claimed compositions would not have been obvious to a person of ordinary skill in the art over Schmaus or Lutz in view of Wang. For these reasons, Applicants respectfully request reconsideration and withdrawal of the rejections over Schmaus in view of Wang and Lutz in view of Wang.

For these reasons, Applicants respectfully request withdrawal the rejections of claims 42 and 72 as well as dependent claims 43-55 and 73-80, respectively.

CONCLUSION

In view of the foregoing remarks, it is respectfully submitted that each of the pending claims is in condition for allowance, and a Notice of Allowance is earnestly solicited. The Examiner is invited to contact the undersigned attorney with any questions, comments or suggestions relating to the referenced patent application.

This response is being submitted on November 11, 2021 with a fee for an Information Disclosure Statement. In the event the Commissioner determines that any additional fees are

required for any reason, the Commissioner is hereby authorized to deduct said fee from Deposit Account No.: 50-3111/Matter No.: 484799.000.

Respectfully submitted,

Date: November 11, 2021

/Aaron Lukas, Reg.# 59,443/

Aaron S. Lukas, Ph.D.
Reg. No. 59,443
COZEN O'CONNOR
1200 Nineteenth Street N.W.
Washington, D.C. 20036
Tel: (202) 912-4823
Fax: (202) 861-1905

PTO/SB/08a (02-18)
Approved for use through 11/30/2020. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Not for submission under 37 CFR 1.99)

Application Number	16262902
Filing Date	2019-01-30
First Named Inventor	BURGO, Rocco V.
Art Unit	1612
Examiner Name	HOLLOMAN, Nannette
Attorney Docket Number	INLX.0198U2/484799

U.S.PATENTS Remove

Examiner Initial*	Cite No	Patent Number	Kind Code[1]	Issue Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear
	1	7385092	B2	2008-06-10	Osterholt, et al.	
	2	8450543	B2	2008-05-13	Peters, et al.	
	3	9732295	B1	2017-08-15	Wright, et al.	

If you wish to add additional U.S. Patent citation information please click the Add button. Add

U.S.PATENT APPLICATION PUBLICATIONS Remove

Examiner Initial*	Cite No	Publication Number	Kind Code[1]	Publication Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear
	1	20050069997	A1	2005-03-31	Adkesson, et al.	

If you wish to add additional U.S. Published Application citation information please click the Add button. Add

FOREIGN PATENT DOCUMENTS Remove

Examiner Initial*	Cite No	Foreign Document Number[3]	Country Code[2] i	Kind Code[4]	Publication Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear	T[5]
	1	104926600	CN	A	2015-09-23	XUEYI, Hu, et al.		X

INFORMATION DISCLOSURE STATEMENT BY APPLICANT (Not for submission under 37 CFR 1.99)		
	Application Number	16262902
	Filing Date	2019-01-30
	First Named Inventor	BURGO, Rocco V.
	Art Unit	1612
	Examiner Name	HOLLOMAN, Nannette
	Attorney Docket Number	INLX.0198U2/484799

	2	105312044	CN	A	2016-02-10	ZHAOSHENG, Jin, et al.		☒
	3	2008007463	JP	A	2008-01-17	TATSURO, Yamamoto, et al.		☒
	4	2004078336	WO	A2	2004-09-16	ZIEHE, Holgar, et al.		☐
	5	2013077904	WO	A1	2013-05-30	WRIGHT, Michael E.		☐
	6	2013106065	WO	A1	2013-07-18	WRIGHT, Michael E.		☐

If you wish to add additional Foreign Patent Document citation information please click the Add button Add

NON-PATENT LITERATURE DOCUMENTS Remove

Examiner Initials*	Cite No	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc), date, pages(s), volume-issue number(s), publisher, city and/or country where published.	T5
	1	ASTMD6866-05, Standard Test Methods for Determining the Biobased Content of Natural Range Materials Using Radiocarbon and Isotape Ratio Mass Spectrometry Analysis, Mar. 1. 2005	
	2	Lu, Metabolic Engineering 44:77 (Nov 1, 2017)	
	3	International Search Report and Written Opinion of International Application No. PCT1US19/15928 dated Apr. 19, 2019	

If you wish to add additional non-patent literature document citation information please click the Add button Add

INFORMATION DISCLOSURE STATEMENT BY APPLICANT (Not for submission under 37 CFR 1.99)		
	Application Number	16262902
	Filing Date	2019-01-30
	First Named Inventor	BURGO, Rocco V.
	Art Unit	1612
	Examiner Name	HOLLOMAN, Nannette
	Attorney Docket Number	INLX.0198U2/484799

EXAMINER SIGNATURE			
Examiner Signature		Date Considered	

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through a citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

[1] See Kind Codes of USPTO Patent Documents at www.USPTO.GOV or MPEP 901.04. [2] Enter office that issued the document, by the two-letter code (WIPO Standard ST.3). [3] For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. [4] Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. [5] Applicant is to place a check mark here if English language translation is attached.

INFORMATION DISCLOSURE STATEMENT BY APPLICANT (Not for submission under 37 CFR 1.99)		
	Application Number	16262902
	Filing Date	2019-01-30
	First Named Inventor	BURGO, Rocco V.
	Art Unit	1612
	Examiner Name	HOLLOMAN, Nannette
	Attorney Docket Number	INLX.0198U2/484799

CERTIFICATION STATEMENT

Please see 37 CFR 1.97 and 1.98 to make the appropriate selection(s):

That each item of information contained in the information disclosure statement was first cited in any communication from a foreign patent office in a counterpart foreign application not more than three months prior to the filing of the information disclosure statement. See 37 CFR 1.97(e)(1).

OR

☐ That no item of information contained in the information disclosure statement was cited in a communication from a foreign patent office in a counterpart foreign application, and, to the knowledge of the person signing the certification after making reasonable inquiry, no item of information contained in the information disclosure statement was known to any individual designated in 37 CFR 1.56(c) more than three months prior to the filing of the information disclosure statement. See 37 CFR 1.97(e)(2).

See attached certification statement.

The fee set forth in 37 CFR 1.17 (p) has been submitted herewith.

☒ A certification statement is not submitted herewith.

SIGNATURE

A signature of the applicant or representative is required in accordance with CFR 1.33, 10.18. Please see CFR 1.4(d) for the form of the signature.

Signature	/Aaron S. Lukas, Reg.# 59,443/	Date (YYYY-MM-DD)	2021-11-11
Name/Print	Aaron S. Lukas, Ph.D.	Registration Number	59,443

This collection of information is required by 37 CFR 1.97 and 1.98. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 1 hour to complete, including gathering, preparing and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

Privacy Act Statement

The Privacy Act of 1974 (P.L. 93-579) requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C. 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether the Freedom of Information Act requires disclosure of these record s.

2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.

3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.

4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).

5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.

6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).

7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.

8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspections or an issued patent.

9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

(19)中华人民共和国国家知识产权局

(12)发明专利

(10)授权公告号 CN 105312044 B
(45)授权公告日 2018.02.13

(21)申请号 201410353875.X

(22)申请日 2014.07.24

(65)同一申请的已公布的文献号
申请公布号 CN 105312044 A

(43)申请公布日 2016.02.10

(73)专利权人 中国石油化工股份有限公司
地址 100728 北京市朝阳区朝阳门北大街22号
专利权人 中国石油化工股份有限公司上海石油化工研究院

(72)发明人 金照生 李亚男 许茗喆 吕晓渊

(51)Int.Cl.
B01J 23/04(2006.01)
B01J 21/10(2006.01)
C07C 11/02(2006.01)
C07C 1/24(2006.01)

(56)对比文件
Behrooz Fallah等.A New Nano-(2Li2O/MgO) Catalyst/Porous Alpha-Alumina Composite for the Oxidative Coupling of Methane Reaction.《AIChE Journal》.2009,第56卷(第3期),第717-728页.

审查员 程远梅

权利要求书1页 说明书5页

(54)发明名称

C6～C10醇脱水制备相应碳数烯烃的催化剂

(57)摘要

本发明涉及C_6～C_{10}醇脱水制备相应碳数烯烃的催化剂，C_6～C_{10}醇脱水制备相应碳数烯烃方法，主要解决现有技术中存在脱水催化剂稳定性差的问题，本发明通过采用C_6～C_{10}醇脱水制备相应碳数烯烃的催化剂，所述催化剂以重量份数计，包括以下组份：a)0.01～30份的IA或ⅡA中的金属氧化物至少其中之一；b)70～99.99份的Al_2O_3；所述金属氧化物与Al_2O_3的份数之和为100份的技术方案，较好地解决了该技术问题，可用于C_6～C_{10}醇脱水制备相应碳数烯烃工业生产中。

CN 105312044 B

1.C_6～C_{10}醇脱水制备相应碳数烯烃的方法，包括在所述催化剂存在下使所述醇或所述醇的含水混合物为反应原料进行脱水反应生成所述相应碳数烯烃；

所述催化剂以重量份数计，包括以下组份：

a)0.01～30份的金属氧化物，所述金属氧化物为Li_2O和MgO；

b)70～99.99份的Al_2O_3；

所述金属氧化物与Al_2O_3的份数之和为100份。

2.根据权利要求1所述的方法，其中，以重量份数计，所述金属氧化物的用量为0.05～20份，Al_2O_3的用量为80～99.95份。

3.根据权利要求2所述的方法，其中，以重量份数计，所述金属氧化物的用量为0.1～15份，Al_2O_3的用量为85～99.9份。

4.根据权利要求1所述的方法，其中，所述的醇为1-辛醇，所述的相应碳数烯烃为1-辛烯。

5.根据权利要求1或4所述的方法，其中，所述醇的含水混合物中醇的重量百分浓度为≥5%且＜100%。

6.根据权利要求1或4所述的方法，其中，反应温度为200～500℃。

7.根据权利要求1或4所述的方法，其中，以醇计反应原料体积空速为0.1～25小时$^{-1}$。

8.根据权利要求1或4所述的方法，其中，所述催化剂的制备方法包括以下步骤：将所需量的选自拟薄水铝石、薄水铝石、三水铝石或湃耳石中的至少一种、含Li和含Mg的化合物、助挤剂和胶溶剂混合，成型，然后干燥、焙烧，得到所述催化剂。

C_6～C_{10}醇脱水制备相应碳数烯烃的催化剂

技术领域

[0001] 本发明涉及C_6～C_{10}醇脱水制备相应碳数烯烃的催化剂，C_6～C_{10}醇脱水制备相应碳数烯烃方法，尤其涉及1-辛醇脱水制备1-辛烯的方法。

背景技术

[0002] 正辛烷是无色透明液体，不溶于水，溶于乙醇、乙醚、苯、丙酮等多数有机溶剂，主要用作高档溶剂和有机合成。

[0003] 正辛烷的可以从相关石油馏分分离得到，由于石油馏分组份复杂，难以获得纯度高的产品。

[0004] 辛醇脱水制辛烯一般采用改性的氧化铝催化剂。

[0005] 黄晴等发表的论文〔2-辛醇气相脱水合成2-辛烯，工业催化，2007年3月，第15 卷，第3期，P50-53〕报道，在330℃，0.35h^{-1}空速下，600小时稳定性试验中2-辛烯收率最高达到93%，但是稳定性不理想，600小时收率降低至75%。

[0006] 高占先等发表等论文〔改性氧化铝催化伯醇脱水合成1-烯烃研究，大连理工大学学报，2001年7月，第41卷，第4期，P412-415〕报道，通过对氧化铝催化剂改性，340℃、常压下，辛醇转化率达到96.8%，辛烯选择性约为70%，未见稳定性情况。

[0007] 辛醇脱水，一般同时存在双键转移现象，产品中主要有1-，2-，3-双键辛烯。在制取正辛烷情况下，这些异构体都可以加氢成为目的产物正辛烷。因此，也可以使用价格较低的混合辛醇原料经过催化脱水制辛烯。

[0008] 1-辛醇脱水制备1-辛烯，1-辛烯加氢制得正辛烷可以获得高纯度正辛烷产品。辛烯加氢可以使用工业上常用的催化剂如Raney Ni，负载Ni，负载Pd，负载Pt催化剂。

[0009] 综上所述，以往技术中采用的脱水催化剂，存在催化剂稳定性差的缺点。

发明内容

[0010] 本发明所要解决的技术问题之一是现有技术中存在脱水催化剂稳定性差的问题，提供一种C_6～C_{10}醇脱水制备相应碳数烯烃的催化剂。该催化剂具有稳定性好的特点。

[0011] 本发明所要解决的技术问题之二是提供采用上述技术问题之一所述的催化剂的C_6～C_{10}醇脱水制备相应碳数烯烃的方法。

[0012] 为解决上述技术问题之一，本发明的技术方案如下：C_6～C_{10}醇脱水制备相应碳数烯烃的催化剂，所述催化剂以重量份数计，包括以下组份：

[0013] a)0.01～30份的IA或ⅡA中的金属氧化物至少其中之一；

[0014] b)70～99.99份的Al_2O_3；

[0015] 所述金属氧化物与Al_2O_3的份数之和为100份。

[0016] 上述技术方案中，以重量份数计，IA或ⅡA中的金属氧化物的用量优选为0.05～20份，Al_2O_3的用量优选为80～99.95份；更进一步，以重量份数计，IA或ⅡA中的金属氧化物的用量更优选为0.1～15份，Al_2O_3的用量更优选为85～99.9份。

[0017] 上述技术方案中，IA金属氧化物优选自Li_2O，ⅡA金属氧化物优选自MgO。所述催化剂更优选同时包括Li_2O和MgO。

[0018] 为解决上述技术问题之二，本发明的技术方案如下：C_6～C_{10}醇脱水制备相应碳数烯烃的方法，包括在上述技术问题之一的技术方案中任一项所述催化剂存在下使所述醇或所述醇的含水混合物为反应原料进行脱水反应生成所述相应碳数烯烃。

[0019] 上述技术方案中，所述的醇优选为1-辛醇，所述的相应碳数烯烃优选为1-辛烯。

[0020] 上述技术方案中，所述醇的含水混合物中醇的重量百分浓度优选为≥5%且＜100%。

[0021] 上述技术方案中，反应温度优选为200～500℃，更优选为200～450℃，进一步优选为 250～450℃。

[0022] 上述技术方案中，以醇计反应原料体积空速优选为0.1～25小时$^{-1}$，更优选为0.5～20 小时$^{-1}$，进一步优选为0.5～15小时$^{-1}$。

[0023] 上述技术方案中，所述催化剂的制备方法没有特别限制，所述催化剂的优选制备方法包括以下步骤：将所需量的选自拟薄水铝石、薄水铝石、三水铝石或湃耳石中的至少一种、含IA或ⅡA中至少一种金属的化合物、助挤剂和胶溶剂混合，成型，然后干燥、焙烧，得到所述催化剂。

[0024] 所述助挤剂优选方案为选自田菁粉、甘油、石墨、干淀粉或柠檬酸中至少一种。所述胶溶剂优选方案为选自硝酸、柠檬酸、乙二酸或乙酸中的至少一种。所述含IA或ⅡA 金属化合物可以是无机或者有机化合物，但以可以在较高温度下分解而不残留其他离子为适宜，优选方案为选自硝酸盐或乙酸盐中的至少一种。

[0025] 本发明中催化剂的寿命判断依据是经过相同时间反应后，通过差热分析得到催化剂的积碳总量，再经过计算，得到单位时间催化剂的积碳百分比量，即积碳速率(单位：%/小时)。该积碳速率越大，催化剂对应的失活速率就越快，催化剂的寿命就越短。

[0026] 本发明通过添加IA或ⅡA中至少一种金属氧化物，用于C_6～C_{10}醇脱水制备相应碳数烯烃过程时，催化剂的积碳速率与氧化铝相比，大幅度降低了积碳速率，从而使得催化剂具有较好的稳定性和寿命。例如通过同比，在用于1-辛醇脱水制1-辛烯时，本发明催化剂的积碳速率可抵达0.0351%/小时，1-辛醇转化率高达99.53%，1-辛烯选择性高达 98.76%，而氧化铝的积碳速率同比高达0.0874%/小时，1-辛醇转化率高达95.24%，1-辛烯选择性高达93.83，取得了较好的技术效果，可用于C_6～C_{10}醇脱水制备相应碳数烯烃工业生产中。

[0027] 下面通过实施例对本发明作进一步阐述。

具体实施方式

[0028] 【实施例1】

[0029] 称量拟薄水铝石200克于捏合机中，加入2克田菁粉捏合30分钟，滴加0.5w%乙酸锂水溶液60毫升，再加200毫升0.5w%硝酸水溶液捏合30分钟，挤条成型，于120℃烘干12小时，550℃焙烧10小时，即得长为5毫米直径为3毫米催化剂催化剂A。催化剂中Al_2O_3的重量份数为99.95份，Li_2O重量份数为0.05份。为便于比较，将催化剂的组成列于表1。

[0030] 催化剂的性能评价在固定床管式反应器(Φ25×500毫米不锈钢)上进行，催化剂装载量为10毫升，反应时间为100小时。反应产物经气液分离后采用Agilent4890气相色谱

仪分别分析。反应条件及反应结果见表2。

[0031] 【实施例2】

[0032] 称量拟薄水铝石200克于捏合机中，加入2克甘油捏合30分钟，滴加20w％乙酸锂水溶液60毫升，再加200毫升0.5w％柠檬酸水溶液捏合30分钟，挤条成型，于150℃烘干10小时，700℃焙烧5小时，即得长为5毫米直径为3毫米催化剂B。催化剂中Al_2O_3的重量份数为98.08份，Li_2O重量份数为1.92份。为便于比较，将催化剂的组成列于表 1。

[0033] 催化剂的性能评价在固定床管式反应器(Φ25×500毫米不锈钢)上进行，催化剂装载量为10毫升，反应时间为100小时。反应产物经气液分离后采用Agilent4890气相色谱仪分别分析。反应条件及反应结果见表2。

[0034] 【实施例3】

[0035] 称量拟薄水铝石200克于捏合机中，加入2克石墨捏合30分钟，滴加15w％乙酸锂水溶液10毫升，再加200毫升0.5w％乙二酸水溶液捏合30分钟，挤条成型，于140℃烘干10小时，650℃焙烧15小时，即得长为5毫米直径为3毫米催化剂C。催化剂中 Al_2O_3的重量份数为99.76份，Li_2O重量份数为0.24份。为便于比较，将催化剂的组成列于表1。

[0036] 催化剂的性能评价在固定床管式反应器(Φ25×500毫米不锈钢)上进行，催化剂装载量为10毫升，反应时间为100小时。反应产物经气液分离后采用Agilent4890气相色谱仪分别分析。反应条件及反应结果见表2。

[0037] 【实施例4】

[0038] 称量拟薄水铝石200克于捏合机中，加入2克田菁粉捏合30分钟，滴加15w％硝酸锂水溶液50毫升，再加200毫升0.5w％硝酸水溶液捏合30分钟，挤条成型，于120℃烘干12小时，550℃焙烧10小时，即得长为5毫米直径为3毫米催化剂D。催化剂中 Al_2O_3的重量份数为98.85份，Li_2O重量份数为1.15份。为便于比较，将催化剂的组成列于表1。

[0039] 催化剂的性能评价在固定床管式反应器(Φ25×500毫米不锈钢)上进行，催化剂装载量为10毫升，反应时间为100小时。反应产物经气液分离后采用Agilent4890气相色谱仪分别分析。反应条件及反应结果见表2。

[0040] 【实施例5】

[0041] 称量薄水铝石200克于捏合机中，加入2克田菁粉捏合30分钟，滴加15w％硝酸锂水溶液40毫升，再加200毫升0.5w％硝酸水溶液捏合30分钟，挤条成型，于90℃烘干 20小时，500℃焙烧20小时，即得长为5毫米直径为3毫米催化剂E。催化剂中Al_2O_3的重量份数为99.12份，Li_2O重量份数为0.88份。为便于比较，将催化剂的组成列于表 1。

[0042] 催化剂的性能评价在固定床管式反应器(Φ25×500毫米不锈钢)上进行，催化剂装载量为10毫升，反应时间为100小时。反应产物经气液分离后采用Agilent4890气相色谱仪分别分析。反应条件及反应结果见表2。

[0043] 【实施例6】

[0044] 称量薄水铝石200克于捏合机中，加入2克干淀粉捏合30分钟，滴加15w％硝酸锂水溶液40毫升，再加200毫升1.5w％柠檬酸水溶液捏合30分钟，挤条成型，于120℃烘干12小时，600℃焙烧10小时，即得长为5毫米直径为3毫米催化剂F。催化剂中Al_2O_3的重量份数为99.61份，Li_2O重量份数为0.39份。为便于比较，将催化剂的组成列于表 1。

[0045] 催化剂的性能评价在固定床管式反应器(Φ25×500毫米不锈钢)上进行，催化剂

装载量为10毫升，反应时间为100小时。反应产物经气液分离后采用Agilent4890气相色谱仪分别分析。反应条件及反应结果见表2。

[0046] 【实施例7】

[0047] 称量三水铝石200克于捏合机中，加入3克石墨捏合30分钟，滴加15w％硝酸锂水溶液40毫升，再加200毫升1w％乙二酸水溶液捏合30分钟，挤条成型，于100℃烘干 15小时，700℃焙烧5小时，即得长为5毫米直径为3毫米催化剂G。催化剂中Al_2O_3的重量份数为98.83份，Li_2O重量份数为1.17份。为便于比较，将催化剂的组成列于表 1。

[0048] 催化剂的性能评价在固定床管式反应器(Φ25×500毫米不锈钢)上进行，催化剂装载量为10毫升，反应时间为100小时。反应产物经气液分离后采用Agilent4890气相色谱仪分别分析。反应条件及反应结果见表2。

[0049] 【实施例8】

[0050] 同【实施例7】，只是采用硝酸镁。

[0051] 称量三水铝石200克于捏合机中，加入3克石墨捏合30分钟，滴加10w％硝酸镁水溶液48.4毫升，再加200毫升1w％乙二酸水溶液捏合30分钟，挤条成型，于100℃烘干15小时，700℃焙烧5小时，即得长为5毫米直径为3毫米催化剂为H。催化剂中Al_2O_3的重量份数为98.83份，MgO重量份数为1.17份。为便于比较，将催化剂的组成列于表 1。

[0052] 催化剂的性能评价在固定床管式反应器(Φ25×500毫米不锈钢)上进行，催化剂装载量为10毫升，反应时间为100小时。反应产物经气液分离后采用Agilent4890气相色谱仪分别分析。反应条件及反应结果见表2。

[0053] 【实施例9】

[0054] 称量三水铝石200克于捏合机中，加入3克石墨捏合30分钟，滴加15w％硝酸锂水溶液20毫升和10w％硝酸镁水溶液24毫升，再加200毫升1w％乙二酸水溶液捏合30 分钟，挤条成型，于100℃烘干15小时，700℃焙烧5小时，即得长为5毫米直径为3 毫米催化剂I。催化剂中Al_2O_3的重量份数为98.83份，Li_2O重量份数为0.59份，MgO 重量份为0.58份。为便于比较，将催化剂的组成列于表1。

[0055] 催化剂的性能评价在固定床管式反应器(Φ25×500毫米不锈钢)上进行，催化剂装载量为10毫升，反应时间为100小时。反应产物经气液分离后采用Agilent4890气相色谱仪分别分析。反应条件及反应结果见表2。

[0056] 【对比例1】

[0057] 称量拟薄水铝石200克于捏合机中，加入2.5克干淀粉捏合30分钟，加200毫升0.5w％硝酸水溶液捏合30分钟，挤条成型，于120℃烘干12小时，550℃焙烧10小时，即得长为5毫米直径为3毫米催化剂J。催化剂中Al_2O_3的重量份数为100份。为便于比较，将催化剂的组成列于表1。

[0058] 催化剂的性能评价在固定床管式反应器(Φ25×500毫米不锈钢)上进行，催化剂装载量为10毫升，反应时间为100小时。反应产物经气液分离后采用Agilent4890气相色谱仪分别分析。反应条件及反应结果见表2。

[0059] 表1催化剂组成(重量份)

[0060]

	Al_2O_3	Li_2O	MgO

实施例1	99.95	0.05	
实施例2	98.08	1.92	
实施例3	99.76	0.24	
实施例4	98.85	1.15	
实施例5	99.12	0.88	
实施例6	99.61	0.39	
实施例7	98.83	1.17	
实施例8	98.83		1.17
实施例9	98.83	0.59	0.58
对比例1	100		

[0061] 表2

[0062]

	原料	温度 ℃	空速 小时$^{-1}$	辛醇 转化率%	辛烯 选择性%	积碳速率 %/小时
实施例1	辛醇	330	0.8	100.00	97.82	0.0380
实施例2	35%辛醇	300	0.5	100.00	98.23	0.0419
实施例3	25%辛醇	290	5	99.99	97.19	0.0454
实施例4	45%辛醇	260	12	98.00	97.25	0.0370
实施例5	85%辛醇	280	1	99.96	97.32	0.0400
实施例6	50%辛醇	400	10	99.98	97.03	0.0410
实施例7	75%辛醇	270	6	98.79	98.36	0.0392
实施例8	75%辛醇	270	6	98.92	98.24	0.0382
实施例9	75%辛醇	270	6	99.53	98.76	0.0351
对比例1	75%辛醇	270	6	95.24	93.83	0.0874

[0063] 注:表2中辛醇为1-辛醇;表中原料中辛醇前的百分数表示原料为辛醇和水混合物的情况下原料中辛醇的重量百分比浓度;辛烯为1-辛烯。

Patent Record Full View Friday, May 1 2020 Derwent Innovation

PATENT/PUBLICATION:CN105312044B

Key summary data

Patent	Alive	Publication date	2018-02-13
DWPI family	Alive	Expiration date	2034-07-24 (estimated)
INPADOC family	Alive	Remaining life	5197 days (14 year(s), 2 month(s))
Original assignee	Sinopec Corp., Beijing, 100728, CN, Sinopec Sha...	Domain Influence	2.10
Optimized assignee	CHINA PETROLEUM & CHEMICAL CORP. INC.	Strategic Importance	2.77
Ultimate parent	CHINA PETROLEUM & CHEMICAL CORP. INC.	Combined Patent Impact	2.25

Bibliography

DWPI title

Catalyst used in dehydrogenation of alcohol for producing olefin, comprises specified amount of specified group metal oxide and alumina

Original Title

C6~C10 alcohol dehydration catalyst for preparing corresponding carbon numbers of olefins *(Translation from Clarivate Analytics)*

English Title

Assignee/Applicant

Original: 中国石油化工股份有限公司, **100728** 北京市朝阳区朝阳门北大街**22**号, **CN**
中国石油化工股份有限公司上海石油化工研究院
Sinopec Corp., Beijing, 100728, CN
Sinopec Shanghai Research Institute of Petrochemical Technology

Inventor

金照生,
李亚男,
许茗喆,
吕晓渊,
JIN Zhao-sheng,
LI Ya-nan,
Xu Mingzhe,
LV Xiao-yuan,

Publication date (Kind Code)

2018-02-13 (B)

Application number / Date

CN201410353875A / 2014-07-24

Priority number / Date / Country code

CN201410353875A / 2014-07-24 / CN

DWPI Priority number / Date / Country code

Abstract

Abstract

The invention relates to C6~C10 alcohol dehydration catalyst for preparing corresponding carbon number olefin, C6~C10 alcohol dewatering method preparing corresponding carbon number olefin, mainly solving the problem that the dehydration catalyst stability is bad. The invention uses C6~C10 alcohol dehydration catalyst for preparing corresponding carbon number olefin, the catalyst based on parts by weight, comprising the following components: a) at least one of 0.01~30 parts of I A or II A in the metal oxide, b) 70~99.99 parts of Al2O3; said metal oxide to Al2O3 parts and 100 parts of the technical plan better solves the problem, can be used for C6~C10 alcohol dehydration to prepare corresponding carbon number olefin in industrial production.

(Translation from Clarivate Analytics)

Classes/Indexing

IPC

Current IPC	Invention	Version	Additional	Version
Full	B01J 23/04 B01J 21/10 C07C 1/24 C07C 11/02	20060101 20060101 20060101 20060101	-	-
Main Group	-	-	-	-
Subclass	-	-	-	-

Original IPC	Invention	Version	Additional	Version
Advanced/Full	B01J 23/04 B01J 21/10 C07C 11/02 C07C 1/24	20060101 20060101 20060101 20060101	-	-
Core/Main Group	-	-	-	-

Subclass	-	-	-	-

DWPI manual codes

Expand DWPI manual codes

Legal status

Predictive Analytics

Predictive Type	Data
Probability of Grant	100.00
Probability of Early Lapse	11.80
Probability of Restoration Post-Lapse	-

INPADOC Legal Status

Gazette Date	Code	Description
2018-02-13	GR01 +	PATENT GRANT
2016-03-09	C10	ENTRY INTO SUBSTANTIVE EXAMINATION
2016-02-10	C06 +	PUBLICATION

Get Family Legal Status

Family

Family

INPADOC Family (2) **INPADOC family:** Alive

	Publication number	Publication date	Inventor	Assignee/Applicant	Dead/Alive	Title
	CN105312044B	2018-02-13	JIN Zhao-sheng	Sinopec Corp.		C6～C10醇脱水制备相应碳数烯烃的催化剂
	CN105312044A	2016-02-10	JIN Zhao-sheng	CHINA PETROLEUM & CHEMICAL		Catalyst used for preparing olefin with corresponding carbon number via C6-C10 alcohol dehydration

Claims

Claims

All Claims (8)

Claims (English)

1.C6~C10 alcohol dehydration method, comprising for preparing corresponding carbon number olefin in the presence of the catalyst the aqueous mixture of the alcohol or the alcohol as reaction raw material to dehydration reaction to generate the respective carbon numbers of olefins, the catalyst based on parts by weight. it comprises the following components: a) 0.01~30 parts of metal oxide, the metal oxide is Li2O and MgO, b) 70~99.99 parts of Al2O3, Al2O3 parts of the metal oxide and is 100 parts.

2. The method according to claim 1, wherein, by weight, the dosage of said metal oxide is 0.05~20 parts; the dosage of the Al2O3 is 80~99.95 parts.

3. The method according to claim 2, wherein, by weight, the dosage of said metal oxide is 0.1~15 parts; the dosage of the Al2O3 is 85~99.9 parts.

4. The method according to claim 1, wherein said alcohol is 1-octanol, the corresponding carbon number olefin is 1-octylene.

5. The method according to claim 1 or 4, wherein the weight percentage concentration of the alcohol in the aqueous mixture of the alcohol is more than or equal to 5% and <100 %.

6. The method according to claim 1 or 4, wherein the reaction temperature is 200~500 degrees centigrade.

7. The method according to claim 1 or 4, wherein alcohol reaction raw material volume space velocity is 0.1~25 -1.

8. The method according to claim 1 or 4, wherein the preparation method of the catalyst comprises the following steps: the required amount selected from pseudo-boehmite, boehmite, trihydrate aluminium or Enpai-Otolith at least one of the compound containing, extrusion assistant

and peptizing agent of Li and Mg-containing mixing, moulding, and then drying, roasting to obtain the catalyst.
(Translation from Clarivate Analytics)

Description

Background/ Summary

Background/Summary

Drawing description

Drawing Description

Description

Description

the technical field

The invention relates to C6~C10 alcohol dehydration catalyst for preparing corresponding carbon number olefin, C6~C10 alcohol dewatering method preparing corresponding carbon number olefin, especially 1-octanol dehydration preparation method of 1-octylene.

the background technology

octane is colourless transparent liquid, insoluble in water, soluble in many organic solvent of ethanol, aether, benzene, acetone and so on, mainly used as solvent and organic synthesis.

octane can be separated from associated petroleum fractions obtained, the petroleum fraction component is complex, it is difficult to obtain the high purity of the product.

octanol octene generally uses the modified alumina catalyst.

such as published thesis [2-octanol phase dehydration industrial catalysis, synthesizing 2-octylene, 2007 December 3, 15 roll, 3, P50-53 report, at 330 degrees centigrade, 0.35h-1 air speed 600-hour stability test in 2-octylene yield up to 93%, but stability is not ideal, 600 yield is reduced to 75%.

high pre-emptive published thesis modified-alumina catalytic primary alcohol synthetized by dehydration of 1-olefin research, Dalian University of Technology Journal, December 2001, 7, coil 41, first 4, P412-415 report, by modifying the alumina catalyst, 340 degrees centigrade under the normal pressure, octanol conversion rate reaches 96.8%, the octene selectivity is about 70%, there is no stability condition.

octanol, double bond transfer phenomenon generally exist at the same time, the product is mainly with 1 -, 2 -, 3-double bond octene. under the condition of producing octane, these isomers can be hydrogenated to form target product octane. Therefore, it also can use the mixed raw material with lower price through catalytic dehydrating octene.

1-octanol is dehydrated to prepare 1-octylene, 1-octene hydrogenation octane can obtain high purity n-octane product. octene hydrogenation using a catalyst such as Raney Ni, load Ni, Pd load common in industry, supported Pt catalyst.

In summary, in the prior art the dehydration catalyst, catalyst stability is poor.

invention content

The invention solves the technical problem in the existing technology has bad dehydration catalyst stability problem, to provide a C6~C10 alcohol dehydration catalyst for preparing corresponding carbon number olefins. The catalyst has good stability.

the second technical problem to be solved by the present invention is to provide uses C6~C10 alcohol above technical problem of the catalyst dehydration method of preparing corresponding carbon number olefins.

In order to solve said technical problem, the technical solution of the invention is as follows: C6~C10 alcohol dehydration catalyst for preparing corresponding carbon number olefin, the catalyst based on parts by weight, comprising the following components:

at least one of a) 0.01~30 parts of I A or II A in the metal oxide;

b) 70~99.99 parts of Al2O3;

the metal oxide and Al2O3 parts and 100 parts.

In the technical solution, preferably in an amount of 0.05~20 parts of metal oxide by weight, I A or II A, the dosage of Al2O3 is preferably 80~99.95, further, the dosage of metal oxide by weight, I A or II A is more preferably 0.1~15, more preferably 85~99.9 of dosage of the Al2O3.

In the technical solution, I A metal oxide is preferably selected from Li2O, metal oxide is preferably selected from MgO. the catalyst more preferably comprises Li2O and MgO.

In order to solve said technical problem, the technical solution of the invention is as follows: C6~C10 alcohol dehydration to prepare corresponding carbon number olefins, comprising one of said technical problem the technical solution of any one of the aqueous mixture of the catalyst in the presence of the alcohol or the alcohol as reaction raw material to dehydration reaction to generate the respective carbon numbers of olefins.

In the technical solution, the alcohol is preferably octanol, the corresponding carbon number olefin preferably 1-octene.

In the technical solution, the weight percentage concentration of the alcohol water mixture of said alcohol is preferably equal to 5% and <100 %.

In the technical solution, the reaction temperature is preferably 200~500 degrees centigrade, more preferably 200~450 degrees centigrade, more preferably 250~450 degrees centigrade.

the technical solution in the alcohol reaction volume airspeed of the raw material is preferably 0.1~25 to -1, more preferably 0.5~20 -1, further preferably 0.5~15 to -1.

In the technical solution, the preparation method of the catalyst is not particularly limited, the preferable preparation method of the catalyst comprises the following steps: the required amount selected from pseudo-boehmite, boehmite, trihydrate aluminium or Enpai-Otolith at least one of the containing compound, extrusion assistant and peptizing agent of at least one metal of I A or II A in mixing, moulding, and then drying, roasting to obtain the catalyst.

the extrusion solution preferably is selected from sesbania powder, glycerol, graphite, dry starch or citric acid in at least one kind of. the glue solvent solution is preferably selected from nitric acid, citric acid, at least one oxalic acid or acetic acid. the I A or II A metal compound can be inorganic or organic compounds, but can decompose without leaving other ion is suitable at the higher temperature, the preferable solution is selected from nitrate or acetate of at least one.

judging the life of catalyst according to the invention, after the same time reaction to obtain the catalyst by differential thermal analysis of the carbon amount, and then calculating the percentage amount of unit time to obtain carbon catalyst, namely carbon rate (unit: % per hour). the deposition rate is larger, the deactivation rate catalyst corresponding to the faster, life of the catalyst becomes short.

The invention is added with at least one metal oxide I A or II A in C6~C10 alcohol dehydration for preparing corresponding carbon number olefin process, the deposition rate of the catalyst compared with alumina, which greatly reduces the deposition rate, so that the catalyst has good stability and life. for example, by comparing, used in 1-octanol by dehydrating 1-octylene, the deposition rate of the catalyst of the invention can reach 0.0351%/h, 1-octanol conversion is up to 99.53%, 1-octene selectivity 98.76%, and the deposit rate of alumina up to 0.0874%/hour, 1-octanol conversion as high as 95.24%, as high as 93.83 1-octene selectivity, has good technical effect, can be used for C6~C10 alcohol dehydration to prepare corresponding carbon number olefin in industrial production.

under the present invention is further illustrated by the examples.

Specific execution examples

" embodiment 1 "

weighing 200 of pseudo-boehmite in kneader, adding 2 powder and kneading for 30, dropping 0.5w 60 % acetate water solution, then adding 200 0.5w % nitric acid aqueous solution for 30 minutes, extruding and moulding, roasting for 10 hours at 120 degrees centigrade, drying for 12 hours at 550 degrees centigrade to obtain length of 5 diameter is 3 mm. the catalyst A. Al2O3 in the catalyst by weight, 99.95 parts of Li2O by weight is 0.05 parts. to facilitate comparison, the composition of the catalyst are listed in Table 1.

the performance evaluation of the catalyst on the fixed bed tubular reactor (ϕ 25 * 500 stainless steel), catalyst loading amount is 10, the reaction time is 100 hours. the reaction product after gas-liquid separation using Agilent4890 gas-phase chromatograph analysis. reaction condition and reaction result are shown in table 2.

" embodiment 2 "

weighing 200 of pseudo-boehmite in kneader, kneading for 30 adding 2 of glycerol, adding 20w % acetate aqueous solution 60, then adding 200 0.5w % of citric acid aqueous solution for 30 minutes, extruding and forming, roasting for 5 hours at 150 degrees centigrade, drying for 10 hours at 700 degrees centigrade to obtain length of 5 diameter is 3 mm. the catalyst B. Al2O3 in the catalyst by weight, 98.08 parts of Li2O by weight is 1.92 parts. to facilitate comparison, the composition of the catalyst are listed in Table 1.

the performance evaluation of the catalyst on the fixed bed tubular reactor (ϕ 25 * 500 stainless steel), catalyst loading amount is 10, the reaction time is 100 hours. the reaction product after gas-liquid separation using Agilent4890 gas-phase chromatograph analysis. reaction condition and reaction result are shown in table 2.

" embodiment 3 "

weighing 200 of pseudo-boehmite in kneader, kneading for 30 adding 2 of graphite, 15w dropping 10 % acetate water solution, then adding 200 0.5w % oxalic aqueous solution and kneading for 30, extruding and forming, roasting for 15 hours at 140 degrees centigrade, drying for 10 hours at 650 degrees centigrade to obtain the length of 5 diameter is 3 catalyst. Al2O3 in the catalyst by weight, 99.76 parts of Li2O by weight is 0.24 parts. to facilitate comparison, the composition of the catalyst are listed in Table 1.

the performance evaluation of the catalyst on the fixed bed tubular reactor (ϕ 25 * 500 stainless steel), catalyst loading amount is 10, the reaction time is 100 hours. the reaction product after gas-liquid separation using Agilent4890 gas-phase chromatograph analysis. reaction condition and reaction result are shown in table 2.

" embodiment 4 "

weighing 200 of pseudo-boehmite in kneader, adding 2 powder and kneading for 30, dropping 15w 50 % nitrate water solution, then adding 200 0.5w % nitric acid aqueous solution for 30 minutes, extruding and moulding, roasting for 10 hours at 120 degrees centigrade, drying for 12 hours at 550 degrees centigrade to obtain length of 5 diameter is 3 mm. the catalyst D. Al2O3 in the catalyst by weight, 98.85 parts of Li2O by weight is 1.15 parts. to facilitate comparison, the composition of the catalyst are listed in Table 1.

the performance evaluation of the catalyst on the fixed bed tubular reactor (ϕ 25 * 500 stainless steel), catalyst loading amount is 10, the reaction time is 100 hours. the reaction product after gas-liquid separation using Agilent4890 gas-phase chromatograph analysis. reaction condition and

reaction result are shown in table 2.

" embodiment 5 "

weighing 200 of boehmite in the kneader, adding 2 powder and kneading for 30, dropping 15w 40 % nitrate water solution, then adding 200 0.5w % nitric acid aqueous solution for 30 minutes, extruding and moulding, roasting for 20 hours at 90 degrees centigrade, drying for 20 hours at 500 degrees centigrade to obtain length of 5 diameter is 3 mm. the catalyst E. Al2O3 in the catalyst by weight, 99.12 parts of Li2O by weight is 0.88 parts. to facilitate comparison, the composition of the catalyst are listed in Table 1.

the performance evaluation of the catalyst on the fixed bed tubular reactor (ϕ 25 * 500 stainless steel), catalyst loading amount is 10, the reaction time is 100 hours. the reaction product after gas-liquid separation using Agilent4890 gas-phase chromatograph analysis. reaction condition and reaction result are shown in table 2.

" embodiment 6 "

weighing 200 of boehmite in kneader, kneading for 30 adding 2 of dry starch, adding 15w % 40 nitrate solution, then adding 200 1.5w % of citric acid aqueous solution for 30 minutes, extruding and forming, roasting for 10 hours at 120 degrees centigrade, drying for 12 hours at 600 degrees centigrade to obtain length of 5 diameter is 3 mm. the catalyst F. Al2O3 in the catalyst by weight, 99.61 parts of Li2O by weight is 0.39 parts. to facilitate comparison, the composition of the catalyst are listed in Table 1.

the performance evaluation of the catalyst on the fixed bed tubular reactor (ϕ 25 * 500 stainless steel), catalyst loading amount is 10, the reaction time is 100 hours. the reaction product after gas-liquid separation using Agilent4890 gas-phase chromatograph analysis. reaction condition and reaction result are shown in table 2.

" embodiment 7 "

weighing 200 of gibbsite in kneader, kneading for 30 adding 3 of graphite, 15w dropping 40 % nitrate water solution, then adding 200 1w % oxalic aqueous solution and kneading for 30, extruding and forming, roasting for 5 hours at 100 degrees centigrade, drying for 15 hours at 700 degrees centigrade to obtain length of 5 diameter is 3 mm. the catalyst G. Al2O3 in the catalyst by weight, 98.83 parts of Li2O by weight is 1.17 parts. to facilitate comparison, the composition of the catalyst are listed in Table 1.

the performance evaluation of the catalyst on the fixed bed tubular reactor (ϕ 25 * 500 stainless steel), catalyst loading amount is 10, the reaction time is 100 hours. the reaction product after gas-liquid separation using Agilent4890 gas-phase chromatograph analysis. reaction condition and reaction result are shown in table 2.

" embodiment 8 "

with the " embodiment 7 ", only uses magnesium nitrate.

weighing 200 of gibbsite in kneader, kneading for 30 adding 3 of graphite, dropping 10w % magnesium nitrate water solution 48.4, then adding 200 1w % oxalic aqueous solution and kneading for 30, extruding and forming, roasting for 5 hours at 100 degrees centigrade, drying for 15 hours at 700 degrees centigrade to obtain length of 5 diameter is 3 mm. the catalyst H. Al2O3 in the catalyst by weight is 98.83 parts, MgO 1.17 shares of weight. to facilitate comparison, the composition of the catalyst are listed in Table 1.

the performance evaluation of the catalyst on the fixed bed tubular reactor (ϕ 25 * 500 stainless steel), catalyst loading amount is 10, the reaction time is 100 hours. the reaction product after gas-liquid separation using Agilent4890 gas-phase chromatograph analysis. reaction condition and reaction result are shown in table 2.

" embodiment 9 "

weighing 200 of gibbsite in kneader, kneading for 30 adding 3 of graphite, adding 15w % nitrate water solution and 20 of 10w wt % magnesium nitrate water solution 24, then adding 200 1w % oxalic aqueous solution and kneading for 30, extruding and forming, roasting for 5 hours at 100 degrees centigrade, drying for 15 hours at 700 degrees centigrade to obtain length of 5 diameter is 3 mm. the catalyst I. Al2O3 in the catalyst by weight, 98.83 parts of Li2O by weight, 0.59 parts of MgO by weight part is 0.58 parts. to facilitate comparison, the composition of the catalyst are listed in Table 1.

the performance evaluation of the catalyst on the fixed bed tubular reactor (ϕ 25 * 500 stainless steel), catalyst loading amount is 10, the reaction time is 100 hours. the reaction product after gas-liquid separation using Agilent4890 gas-phase chromatograph analysis. reaction condition and reaction result are shown in table 2.

"Comparative Example 1"

weighing 200 of pseudo-boehmite in kneader, kneading for 30 adding 2.5 of dry starch, adding 200 0.5w % nitric acid aqueous solution for 30 minutes, extruding and moulding, roasting for 10 hours at 120 degrees centigrade, drying for 12 hours at 550 degrees centigrade to obtain the length of 5 diameter is 3 mm. the catalyst. Al2O3 in the catalyst by weight is 100 parts. to facilitate comparison, the composition of the catalyst are listed in Table 1.

the performance evaluation of the catalyst on the fixed bed tubular reactor (ϕ 25 * 500 stainless steel), catalyst loading amount is 10, the reaction time is 100 hours. the reaction product after gas-liquid separation using Agilent4890 gas-phase chromatograph analysis. reaction condition and reaction result are shown in table 2.

Table 1 catalyst composition (parts by weight)

Al2O3Li2OMgO Example 199.950.05 Example 298.081.92 Example 399.760.24 Example 498.851.15 Example 599.120.88 Example 699.610.39 Example 798.831.17 Example 898.831.17 Example 998,830.590.58 Comparative Example 1100

Table 2

note: in Table 2 octanol is 1-octanol, percent octanol in the raw material list represents the weight percentage concentration in octanol raw material is mixture of octanol and water in the octylene is 1-octene.
C6~C10 alcohol dehydration catalyst for preparing corresponding carbon numbers of olefins *(Translation from Clarivate Analytics)*

Citations

Citation *Record level*

Citing Patents (0)

Cited Patents (2)

	Publication number	Publication date	Application date	Inventor	Assignee/Applicant	Title	Relevance	Source
	US20070043182A1	2007-02-22	2005-11-02	Martin Joel L.		Multimodal polyethylene compositions and pipe made from same	A	0 (Examiner)
	CN102190543A	2011-09-21	2010-03-03	YANG Wei-min	CHINA PETROLEUM & CHEMICAL	Method for preparing ethylene by dehydrating ethanol	YX	0 (Examiner)

Cited Non-patents (2)

- Behrooz Fallah and the like. A New Nano - (2Li2O/MgO) Catalyst/porous α-Alumina Composite for the Oxidative m/z Methane Reaction. "AIChE Journal". 2009, volume 56 (3), page 717-728.

* Behrooz Fallah 等.A New Nano-(2Li2O/MgO) Catalyst/Porous Alpha-Alumina Composite for the Oxidative Coupling of Methane Reaction.《AIChE Journal》.2009,第56卷(第3期),第717-728页.

DPCI Citation *Patent family level*

DPCI Citing Patents (0)

DPCI Cited Patents (0)

DPCI Cited Non-patents (0)

DPCI Citation Counts

DPCI Field of search

DPCI Citing Patents Country/Region

DPCI Cited Patents Country/Region

Other

Examiner

Related applications

Parent/Child	Application number	Filed Date	Publication number	Publication date	Type of Relationship	Status
P	CN201410353875A	2014-07-24	CN105312044A	2016-02-10	Related publication	-

Copyright 2007-2020 CLARIVATE ANALYTICS

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
16 September 2004 (16.09.2004)

PCT

(10) International Publication Number
WO 2004/078336 A2

(51) International Patent Classification[7]: B01J

(21) International Application Number: PCT/EP2004/002086

(22) International Filing Date: 2 March 2004 (02.03.2004)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
103 09 367.2 3 March 2003 (03.03.2003) DE

(71) Applicant *(for all designated States except US)*: **SASOL GERMANY GMBH** [DE/DE]; Anckelmannsplatz 1, 20537 Hamburg (DE).

(72) Inventors; and

(75) Inventors/Applicants *(for US only)*: **ZIEHE, Holger** [DE/DE]; Timm-Kröger-Strasse 8, 25524 Itzehoe (DE). **SCHIMANSKI, Jürgen** [DE/DE]; Trischenweg 13, 25541 Brunsbüttel (DE). **BRASCH, Andrea** [DE/DE]; Lütjenmarschweg 156, 25704 Meldorf (DE). **TÖNSEN, Ernst-Otto** [DE/DE]; Hafenstrasse 1, 25541 Brunsbüttel (DE).

(74) Agent: **SCHUPFNER, Georg, U.**; Müller, Schupfner & Gauger, Parkstrasse 1, 21244 Buchholz (DE).

(81) Designated States *(unless otherwise indicated, for every kind of national protection available)*: AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BW, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DK, DM, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NA, NI, NO, NZ, OM, PG, PH, PL, PT, RO, RU, SC, SD, SE, SG, SK, SL, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, YU, ZA, ZM, ZW.

(84) Designated States *(unless otherwise indicated, for every kind of regional protection available)*: ARIPO (BW, GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HU, IE, IT, LU, MC, NL, PL, PT, RO, SE, SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Published:
— *without international search report and to be republished upon receipt of that report*

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: PROCESS FOR THE DEHYDRATION OF ALCOHOLS YIELDING α-OLEFINS

(57) Abstract: The present invention relates to a process for producing a-olefins by dehydration of alcohols in the presence of γ-alumina. Furthermore, this invention relates to the use of γ-alumina as a dehydration catalyst.

WO 2004/078336 A2

PROCESS FOR THE DEHYDRATION OF ALCOHOLS YIELDING α -OLEFINS

The present invention relates to a process for producing α-olefins by dehydration of alcohols in the presence of γ–alumina. Furthermore, this invention relates to the use of γ–alumina as a dehydration catalyst.

Linear α-olefins are produced on a large scale for example by ethylene oligomerisation, Fischer-Tropsch synthesis or, as in the case of 1-hexene, by controlled trimerisation. The ethylene oligomerisation can be performed for example according to Ziegler by a two-stage process, i.e. by a building-up reaction using triethyl aluminium, followed by 'short-time high-temperature displacement' (termed ALFEN process), or by a single-stage high-temperature process with triethyl aluminium catalysis.

Another process for producing α-olefins is the dehydration of alcohols, which has been described in detail for example by Knözinger, H., *Angew. Chem. (Applied Chemistry), Int. Ed.*, vol. 7, 1968, no. 10, p. 791-805.

Short-chain linear α-olefins are especially employed as co-monomers for making polyethylene copolymers. High purity is essential for this use, particularly as regards the 1-olefins content, plus the lowest possible amount of impurities, such as internal, branched, or cyclic alkenes, and dienes or alkines. It is known that few ppm of impurities already result in rapid deactivation of the polymerisation catalyst. Owing to the small boiling-point difference of the double-bond isomers and structural isomers of the olefins, which makes splitting by distillation expensive, the α-selectivity of the dehydration step is most important.

By the term 'α-selectivity' as used herein is meant the ratio of the α-olefin formed to the total olefins formed, i.e. the total of α-olefins, internal, branched, and cyclic olefins.

Suitable alumina catalysts for this application comprise for example zinc- and zirconium-doped aluminas (cf. e.g. U.S. patent 4,260,845; EP patent 0 150 832-B1, respectively). The α-selectivity can also be improved by addition of bases, which, however, has an adverse effect on the reactivity. For example, base-doped alumina

CONFIRMATION COPY

catalysts, such as barium-doped alumina, require significantly higher temperatures than the corresponding undoped catalysts. Acidic aluminas can be used at considerably lower temperatures causing, however, more isomerisation reactions and hence a decrease in α-selectivity.

Furthermore, it is known that there are processes, wherein prior to use the bases are homogeneously mixed with the alcohol. German patent DE 39 15 493 C2 describes the improvement of α-selectivity in the dehydration of fatty alcohols on γ-alumina by addition of 20 to 300 ppm of ammonia. However, it is a disadvantage of said process to add an additional component, namely ammonia, which later must be separated.

The commonly known processes of alumina-catalysed dehydration have the disadvantage of poor α-selectivity obtained with the customary catalysts. The selectivity can be improved at the expense of the turnover or by costly modification of the catalyst or by additon of further substances, which later must be separated. Moreover, reaction temperatures of greater than 350°C are necessary in order to achieve a fairly good conversion.

It is, therefore, an object of the present invention to provide a process for producing α-olefins by dehydration of alcohols, which process yields a high α-selectivity and good conversion at a low reaction temperature of less than 350 °C, particularly 320 °C and lower, without addition of bases or acids or without catalyst doping. Another object of the present invention is to provide a novel dehydration catalyst.

According to the present invention, the problem has been solved by a process for producing α-olefins by dehydration of alcohols in the presence of γ-alumina, wherein the alcohol is brought into contact with at least one γ-alumina having

- a pore volume of greater than 0.9 ml/g, preferably greater than 1.0 ml/g (measured in accordance with DIN 66133, contact angle 131°C),
- at least one pore radii maximum in the mesopores range (10 to 250 Å) from 20 to 90 Å, preferably 30 to 80 Å, and
- at least one additional pore radii maximum in the macropores range (greater than 250 Å).

The dehydration is preferably carried out at temperatures in the range from 260 to 350 °C, most preferably from 280 to 320 °C. The preferred embodiments of the subject invention are set out in the subordinate claims or hereinbelow.

According to the invention, the preferable alcohols are comprised of linear or branched 1-alkanols having 4 to 14 carbon atoms. Examples of suitable 1-alkanols include 1-butanol, 1-pentanol, 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol, 1-decanol, 1-undecanol, 1-dodecanol, 1-tridecanol, 1-tetradecanol, 2-ethyl-1-hexanol, and 2-butyl octanol, of which 1-hexanol and 1-octanol are particularly preferred.

In addition to all-γ-phase alumina, catalyst blends composed of γ- and δ-phases may be used as well. The γ-alumina according to the invention is mostly comprised of the γ-phase, namely, more than 50 wt%, preferably more than 90 wt%, and it has a bimodal pore radii distribution comprising at least one pore radius maximum in the mesopores range (pore radius 10 to 250 Å) and one in the macropores range (pore radius > 250 Å) with a total pore volume of greater than 0.9 ml/g and an amount of mesopores of preferably greater than 0.6 ml/g (determined in accordance with DIN 66133 by the mercury intrusion method at a contact angle of 131°). At least one maximum of the mesopores radii distribution of the γ-alumina of the invention is in the range from 20 to 90 Å, preferably 30 to 80 Å, whereas the preferable maximum of the macropores is greater than 1,000 Å.

It is preferable to use alumina having an Al_2O_3 content of >99 wt%, which unlike the aluminas described in US 4,260,845 or EP 0 150 832-B1, is not doped with zinc or zirconium or other additives, a process-conditioned amount of zinc and zirconium of typically less than 10 ppm notwithstanding. It is furthermore preferable not to employ any other type of dehydration catalysts, including co-catalysts or active carriers.

An example of a typical composition of the catalysts of the invention is given in Table I hereinbelow.

Table I

Catalyst Composition	PURAL™ KR 1	
Al	wt%	53.6
Si	ppm	553
Ca	ppm	176
Na	ppm	159
Fe	ppm	156
Mg	ppm	78
Ga	ppm	70
Ti	ppm	14
Pb	ppm	14
Cr	ppm	11
K	ppm	<10
Li, Zr, B, Mo, Ni, Mn, Zn,Cu,Co	each ≤ 5 ppm	

In principle, any commercially available γ-alumina catalyst can be employed for the dehydration of 1-alcohols, but their use is less advantageous, compared to the catalysts of the invention. The γ-Al_2O_3-catalysts of the invention differ from conventional catalysts in their physical properties, such as surface, porosity, pore geometry, and chemical composition. The catalysts of the invention excel by their high pore volume of greater than 0.9 ml/g and pore radii distribution which is at least bimodal and has at least one maximum in the mesopores range from 30 to 80 Å and one in the macropores range of preferably >1,000 Å, wherein the pore volume of the mesopores preferably makes up more than 40 % of the total pore volume, whereas independently thereof the pore volume of the macropores preferably constitutes more than 20 %. In the dehydration of 1-alcohol the special combination of physical properties of this alumina surprisingly yields a crude product having a significantly higher α-selectivity with unimpaired conversion (cf. Table II).

The γ-alumina catalysts of the invention are different from the conventional catalysts usually employed for the dehydration of 1-alcohols yielding 1-olefins, because they have been modified by the manufacturing method, especially with respect to their physical properties, such that they have turned out to be

surprisingly selective in the dehydration process, particularly as regards the purity of the desired 1-olefin.

The process of the invention is preferably carried out at a pressure ranging from 10 to 2,000 mbar. The high-porosity alumina is preferably employed for example in the form of beads, extrudate clippings, granules, or pellets. The dehydration is preferably carried out continuously, most preferably in a continuous tube reactor.

The process of the invention can be carried out for example as follows:

The preferably gaseous alcohol is passed preferably at 280 to 320 °C through a reactor, especially a tube reaetor, packed with the alumina of the invention. Pressure and temperature are such that at the reaction conditions of choice the catalyst is not contacted by liquid, e.g. the higher-boiling ether, which may theoretically be formed as an intermediate during the reaction. The alcohol may optionally be mixed with inert gas, e.g. nitrogen. The alumina is present for the most part as a g-phase material, preferably in the form of granules, extrudate, or beads.

The reaction product is condensed, the aqueous phase is separated, and the organic phase is analysed by gas chromatography in order to examine the reaction. With the purpose of evaluating the catalyst activity, the reactor is operated at steady conditions until a constant composition/stationary state is reached. The organic phase obtained after phase separation is distilled and both the unreacted alcohol and the dialkyl ether formed can be recycled to the process. Alternatively, the alcohol/dialkyl ether mixture can be separated from the olefin/water mixture by condensation immediately after the reaction, followed by direct recycling to the process.

The dihexyl ether which is usually formed as a by-product during the reaction of 1-hexanol can be distilled off and recycled to the process. The compositions of the crude educts/products and the test conditions for various catalysts have been compiled in Table II .

PURAL™ KR1 granules and extrudates are exemplary of the high-prosity bimodal γ-aluminas of the invention. When comparing the α-selectivities at 300 °C and a feed rate of 2 ml/min, their remarkable superiority over the

Südchemie catalyst is obvious. This superiority is yet more evident at 325 °C. PURAL™ KR2, too, is a high-porosity γ-alumina, but with a monomodal pore radii distribution. The P 180 catalyst is a high-porosity alumina, but compared with the catalyst of the invention, it has a considerably greater average pore radius in the mesopores range.

It has become apparent that the process for producing 1-hexene in the co-monomer mode utilising the alumina according to the invention is much more cost-effective. For example, working-up of the crude product by distillation in order to obtain a 1-hexene content of > 99 % can be accomplished by means of a column with fewer separation stages. The turnover can be affected by temperature, feed quantity (MHSV), and catalyst bed length, but the α-selectivity decreases as the turnover increases.

Figure 1 shows the pore radii distributions of PURAL™ KR1 according to example 3 and of PURAL™ KR2 according to comparative example 4. The pore radii were determined by the Hg intrusion method according to DIN 66133 at a contact angle of 131° using a measuring apparatus of *micromeritics* company.

Figure 2 shows the 1-hexene content in the crude product versus the α-selectivity as a function of feed quantity and temperature. The data points plotted for a 1-hexene content of less than 30 % have been obtained at 300 °C, whereas the data collected for a 1-hexene content of greater than 30 % are based on 325 °C (legend: (1) 300°C, 2 ml/min; (2) 325°C, 3 ml/min; (3) 325°C, 2 ml/min; (4) 325°C, 1 ml/min). With an increasing turnover the differences in the α-selectivities of various catalysts become particularly plain.

Examples

Preparation of γ-Al_2O_3 Extrudates

2 kgs of alumina and 2 kgs of water were mixed for 20 minutes in a Z-type kneader mixer. The resultant paste was extruded through a 2-mm strainer of a single-screw extruder and then was dried and calcined onto the desired surface.

Preparation of γ-Al_2O_3 Granules

Alumina and water were continuously mixed in a double-screw extruder at a feed rate of 15 kgs/h. The resultant granules were dried and then calcined onto the desired surface.

Examples 1 through 5 **Dehydration of Hexanol on Alumina**
(for the specification, see Table II)

In a continuously operated tube reactor (23 mm in diameter, 100 mm in length), 2 ml/min of hexanol vapour having a purity of 99.4 % and 0.25 l/min of nitrogen were passed at 300 °C/325 °C and 2 bar through a 50-mm catalyst bed comprised of 7.2 to 9.9 grams of catalyst, depending on the bulk density.

When using the monomodal γ-alumina of Südchemie (CS 331-1; pore volume 0.82 ml/g) at 300°C reaction temperature, the organic phase obtained after phase separation of the crude product comprised 44.2 % hexanol, 32.2 % dihexyl ether, and 20.7 % 1-hexene (comparative example 4). The α-selectivity was 96.5 %.

When using PURAL™ KR1 at 300°C reaction temperature, the organic phase obtained after phase separation of the crude product comprised 35.2 % hexanol, 38.7 % dihexyl ether, and 24.5 % 1-hexene. The α-selectivity of the hexenes was 97.7 %.

Table II

Gas-Phase Dehydration of 1-Hexanol on Al_2O_3 Catalysts

Examples	1		2		3	4	5
Catalyst Al_2O_3	CS 331-1 (Südchemie)		PURAL KR1 (Sasol)		PURAL KR1 (Sasol)	PURAL KR2 (Sasol)	P180 (Sasol)
Type	Extrudates		Granules		Extrudates	Extrudates	Extrudates
Surface (m^2/g)	235		184		232	215	157
Pore volume (Hg) [ml/g], thereof pore radii of	0.83		1.31		1.28	0.89	0.99
< 300 Å	0.69 (83%)		0.79 (60%)		0.77 (60%)	0.84 (94%)	0.77 (78%)
300 – 1,000 Å	0.04 (5%)		0.05 (4%)		0.03 (2%)	0.01 (2%)	0.03 (3%)
> 1,000 Å	0.09 (11%)		0.47 (36%)		0.48 (38%)	0.04 (4%)	0.19 (19%)
Pore radius maximum > 1,000 Å	1,000		2,500		5,000	-	≈ 5,000 (large distribution)
Pore radius maximum < 1,000 Å	50		68/49		50/35	70/40	117
Temperature [°C]	**300**	**325**	**300**	**325**	**325**	**325**	**325**
Feed quantity [ml/min]	2	2	2	2	2	2	2
Composition [wt%]							
1-Hexanol (educt 99.40)	44.22	24.77	35.21	19.03	21.14	21.83	24.25
Dihexyl ether	32.23	14.78	38.65	18.75	15.40	14.33	13.76
1-Hexene	20.704	52.551	24.517	58.421	58.840	58.653	55.90
cis-2-Hexene	0.518	4.210	0.412	1.839	2.320	2.668	3.49
trans-2-Hexene	0.180	1.483	0.116	0.530	0.826	1.007	1.32
cis-3-Hexene	0.013	0.118	0.010	0.046	0.066	0.085	0.08
trans-3-Hexene	0.011	0.103	0.008	0.037	0.056	0.076	0.19
2-Ethyl-1-butene	0.013	0.015	0.016	0.025	0.024	0.022	n. a.
3-+4-Methyl-1-pentene	0.003	0.009	0.003	0.008	0.009	0.009	n. a.
3-Methyl-2-pentene	0.003	0.012	0.002	0.009	0.008	0.010	n. a.
Total of other hexenes	-	-	-	-	-	-	0.31
Total of C_6 olefins	21.45	58.50	25.08	60.92	62.15	62.53	61.29
Conversion [%]	55.52	75.08	64.58	80.86	78.74	78.04	75.60
α-Selectivity [%]	96.54	89.83	97.74	95.91	94.68	93.80	91.21

Patent Claims

1. A process for producing α-olefins by dehydration of alcohols in the presence of γ-alumina,
characterised in that the alcohol is brought into contact with at least one γ-alumina having
   - a pore volume of greater than 0.9 ml/g,
   - at least one pore radii maximum in the mesopores range from 20 to 90 Å, and
   - at least one pore radii maximum in the macropores range, i.e. greater than 250 Å.

2. The process according to claim 1,
characterised in that the pore volume of the mesopores in ml/g (pore radii from 10 to 250 Å) of the γ-alumina makes up more than 40 % of the total γ-alumina pore volume.

3. A process according to any one of the preceding claims,
characterised in that the pore volume of the macropores in ml/g (pore radii greater than 250 Å) of the γ-alumina makes up more than 20 % of the total γ-alumina pore volume.

4. A process according to any one of the preceding claims,
characterised in that the pore volume of the mesopores (pore radii from 10 to 250 Å) of the γ-alumina is greater than 0.6 ml/g.

5. A process according to any one of the preceding claims,
characterised in that the pore volume of the macropores (pore radii greater than 250 Å) of the γ-alumina is greater than 0.3 ml/g.

6. A process according to any one of the preceding claims,
characterised in that the γ-alumina comprises more than 99 wt% Al_2O_3.

7. A process according to any one of the preceding claims,
characterised in that the dehydration is performed at temperatures ranging from 260 to 350 °C, preferably 280 to 320 °C.

8. The process according to any one of the preceding claims, characterised in that the alcohol employed herein is a linear alcohol, preferably at least one linear alcohol selected from the group consisting of 1-butanol, 1-pentanol, 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol, 1-decanol, 1-undecanol, 1-dodecanol, 1-tridecanol and 1-tetradecanol.

9. A process according to any one of the preceding claims, characterised in that the alcohol employed herein is comprised of one or more 1-alkanol(s) having 4 to 14 carbon atoms, particularly 6 to 8.

10. A process according to any one of the preceding claims, characterised in that the γ-alumina is employed in the form of blanks, preferably as beads, pellets, extrudate, or granules, and independently thereof preferably in a fixed-bed reactor.

11. A process according to any one of the preceding claims, characterised in that the alcohol is in the gas phase when contacted with the γ-alumina.

12. A process according to any one of the preceding claims, characterised in that the dehydration is performed at a pressure of 10 to 2,000 mbar.

13. A process according to any one of the preceding claims, characterised in that the dehydration is continuously performed in a tube reactor.

14. A process according to any one of the preceding claims, characterised in that at least one pore radii maximum is found in the macropores range with pore radii of greater than 1,000 Å.

15. The use of any one of the γ-aluminas claimed in any one of the preceding claims as a dehydration catalyst.

Fig. 1

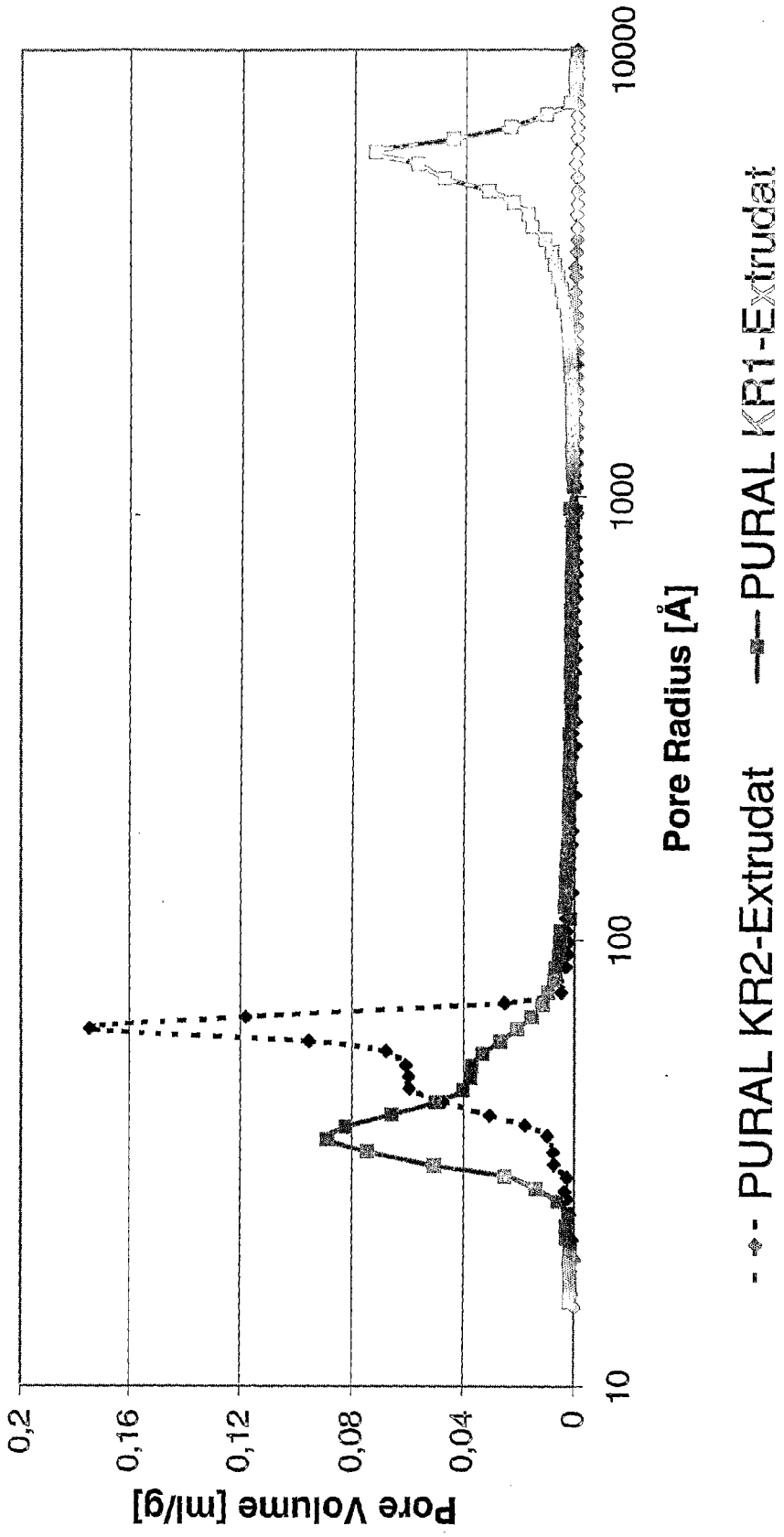
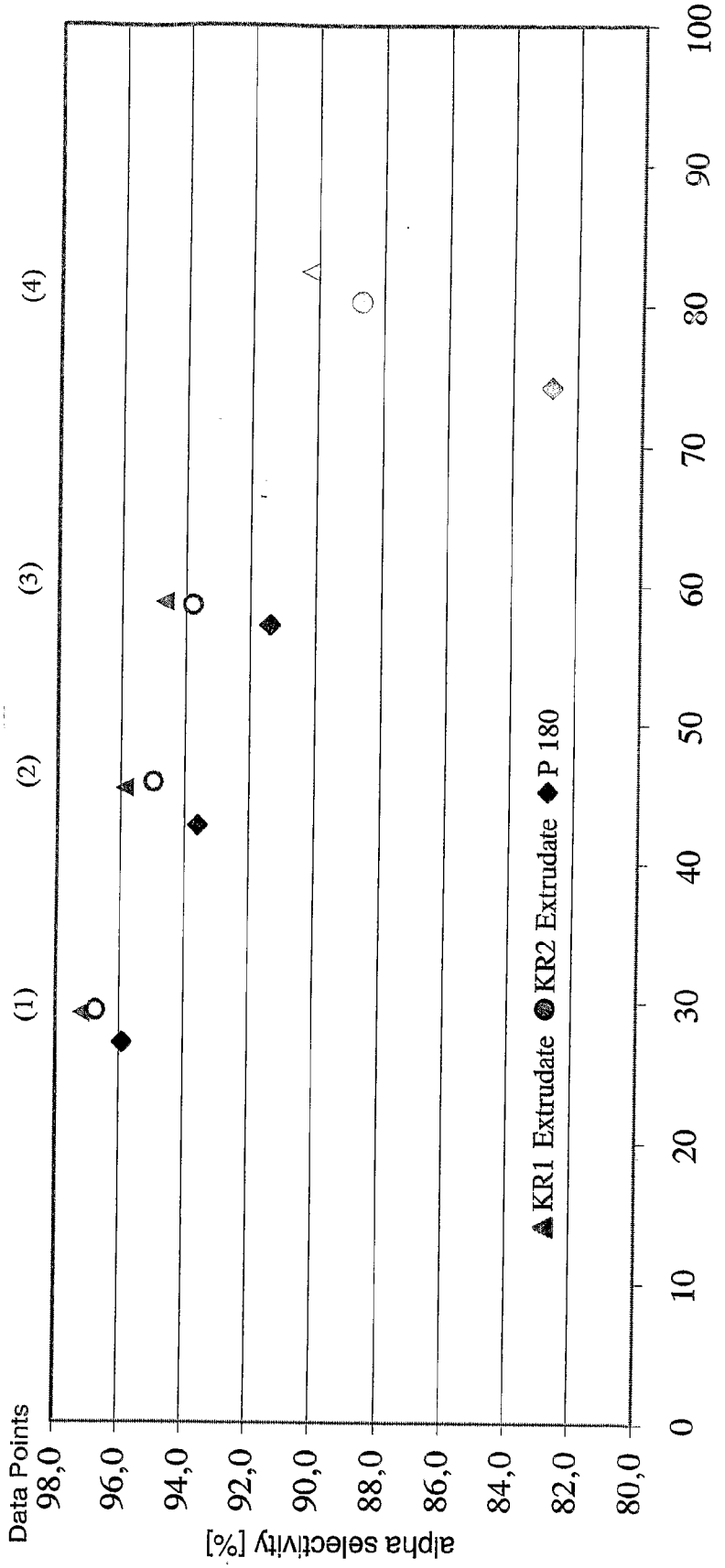

Incremental Pore Volume
Pore Volume [ml/g]
0,2
0,16
0,12
0,08
0,04
0
10
100
1000
10000
Pore Radius [Å]
- PURAL KR2-Extrudat
PURAL KR1-Extrudat

Fig. 2


Data Points
(1)
(2)
(3)
(4)
98,0
96,0
94,0
92,0
90,0
88,0
86,0
84,0
82,0
80,0
alpha selectivity [%]
0
10
20
30
40
50
60
70
80
90
100
1-Hexene Content [%]
KR1 Extrudate
KR2 Extrudate
P 180

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
30 May 2013 (30.05.2013)

WIPO | PCT

(10) International Publication Number
WO 2013/077904 A1

(51) International Patent Classification:
C12P 7/16 (2006.01) *C02F 1/04* (2006.01)
B01D 3/00 (2006.01)

(21) International Application Number:
PCT/US2012/035132

(22) International Filing Date:
26 April 2012 (26.04.2012)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
61/562,231 21 November 2011 (21.11.2011) US
13/433,737 29 March 2012 (29.03.2012) US

(71) Applicant *(for all designated States except US)*: **THE GOVERNMENT OF THE UNITED STATES OF AMERICA AS REPRESENTED BY THE SECRETARY OF THE NAVY** [US/US]; Naval Warfare Center-weapons Division, 1 Administration Circle, Stop 1009, China Lake, CA 93555-6100 (US).

(72) Inventor; and
(75) Inventor/Applicant *(for US only)*: **WRIGHT, Michael, E.** [US/US]; 245 N. Alvord, Ridgecrest, CA 93555 (US).

(74) Agents: **HALEY, Charlene, A.** et al.; Naval Air Warfare Center, Weapsons Division, 1 Administration Circle, China Lake, CA 93555-6100 (US).

(81) Designated States *(unless otherwise indicated, for every kind of national protection available)*: AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, JP, KE, KG, KM, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States *(unless otherwise indicated, for every kind of regional protection available)*: ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Published:

— *with international search report (Art. 21(3))*

(54) **Title**: WATER AND CONTAMINANTS REMOVAL FROM BUTANOL FERMENTATION SOLUTIONS AND/OR BROTHS USING A BRINE SOLUTION

(57) **Abstract**: A process and apparatus are provided for removal of water and other selected contaminants from butanol fermentation broths which can be achieved by continuous extraction with a concentrated salt solution with a regulated pH value. This process greatly reduces distillation energy and losses for the final purification of butanol (normal or iso-). Furthermore, this process allows for generation of sufficiently pure biobutanol (>97%) for use in a subsequent chemical process.

WO 2013/077904 A1

WATER AND CONTAMINANTS REMOVAL FROM BUTANOL FERMENTATION SOLUTIONS AND/OR BROTHS USING A BRINE SOLUTION

STATEMENT REGARDING FEDERALLY SPONSORED RESEARCH OR DEVELOPMENT

[0001] The invention described herein may be manufactured and used by or for the government of the United States of America for governmental purposes without the payment of any royalties thereon or therefor.

FIELD OF THE INVENTION

[0002] The invention generally relates to a fuel and process for dewatering and purifying butanol prepared through a fermentation process.

BACKGROUND

[0003] Purification of butanol from fermentation processes has a long history, nearly as long at the initial discovery of the Acetone-Butanol-Ethanol process by Louis Pasteur in 1862. What has evolved as the most efficient purification process involves some kind of sparge using an inert carry stream. This can be carbon dioxide or simply water. One practiced method takes advantage of the butanol-water azeotrope. This azeotrope, after collection, then phase separates into an organic phase (top) and a bottom water layer. The organic phase is often denoted as the "butanol beer broth," which is decanted away from the bottom water layer. The latter is returned and can be used again as a sparging solvent.

[0004] US Patent Application 12/018,216 summarizes the four general methods of purifying butanol as related to iso-butanol, although the methodology is applicable to both iso- and n-butanol.

[0005] The first method takes the organic layer including butanol and ~15-20 water is simply further purified by fractional distillation. The first fractions are low boiling impurities followed by azeotropic (butanol-water) removal of the water and this is followed by collection of the purified butanol, typically 98% or greater. The butanol-water azeotrope is recycled and must be

distilled again, utilizing process time and energy.

[0006] In method two, the butanol is isolated from the fermentation medium using liquid-liquid extraction in combination with distillation. In this method, the butanol is extracted from the fermentation broth using liquid-liquid extraction with a suitable solvent. The butanol-containing organic phase is then distilled to separate the butanol from the solvent. This method can require large amounts of a flammable co-solvent and presents difficulties with scale.

[0007] In method three, a distillation in combination with adsorption is used to sequester butanol from the fermentation medium. For this process, the fermentation broth containing the butanol is sparged near the azeotropic composition and then the remaining water is removed by use of an adsorbent, such as molecular sieves (Aden et al., Lignocellulosic Biomass to Ethanol Process Design and Economics Utilizing Co-Current Dilute Acid Prehydrolysis and Enzymatic Hydrolysis for Corn Stover, Report NREL/TP-510-32438, National Renewable Energy Laboratory, June **2002**). This can require large amounts of absorbent since butanols typically carry 15-20 wt. % water once physically separated from the water phase.

[0008] A fourth approach is to combine a sparging process to first remove the butanol from the reactor and solids followed by a pervaporation process. As above, the fermentation broth containing the butanol (typically 1.0-2 wt. %) is sparged from the bioreactor at approximately azeotropic composition, after phase separation and the remaining water is removed by pervaporation, for example, through a hydrophilic membrane [Guo et al., *J. Membr. Sci.* 245, 199-210 (**2004**)]. Typically pervaporation processes are difficult for large scale operations and can be extremely costly on a commercial scale. When used before a sparging process and contact is made with the fermentation mixture, biofouling of the membrane is problematic.

BRIEF DESCRIPTION OF THE DRAWINGS

[0001] **FIG 1.** A general overview of process going from sugars (fermentation) to purified biobutanol, according to embodiments of the inventions.

[0002] **FIG 2.** A process flow diagram (PFD) for dewatering and contaminant removal from "biobutanol beer broth" obtained as a sparge from a fermentation vessel, according to embodiments of the inventions.

[0003] **FIG 3**. Proton nuclear magnetic resonance (NMR) spectrum of dewatered bio-1-butanol obtained from an original "butanol beer broth" including 15-18 wt. % water. The original broth was obtained by fermentation of sugars using Clostridium bacteria, according to embodiments of the inventions.

[0004] It is to be understood that the foregoing general description and the following detailed description are exemplary and explanatory only and are not to be viewed as being restrictive of the invention, as claimed. Further advantages of this invention will be apparent after a review of the following detailed description of the disclosed embodiments, which are illustrated schematically in the accompanying drawings and in the appended claims.

DETAILED DESCRIPTION OF EMBODIMENTS OFTHE INVENTION

[0005] The invention generally relates to a process and fuel derived from biobutanol. Biobutanol is butanol that is derived from the fermentation of biomass. It can be as a direct precursor to 1-butene. 1-butene can be utilized to make drop-in JP-5 alternative fuels. Thus, improvements in isolating biobutanol are expected to decrease cost of the alternative JP-5 jet fuel.

[0006] Currently energy intense distillation methods are used to remove water from the biobutanol (as produced in fermentations). A rapid and quick drying with a brine solution can reduce the waste and cost of the associated lengthy distillation process and lead to increased yield of biobutanol.

[0007] Currently distillation alone is used to purify biobutanol on large scale production processes. This is done to remove ethanol, acetone, the predominance of water (~15%), and other minor impurities. By using azeotropic distillation of the butanol followed by, physical separation, and then drying by exposure to a brine solution (saturated sodium chloride or other salts), an almost continuous distillation process can be used and eliminate the primary impurities of the biobutanol.

[0008] An aspect of the invention relates to a purification process of isolating biobutanol including removing biobutanol from a fermentation broth using at least one separation process and phase separating an organic layer and a concentrated biobutanol, dewatering the

concentrated biobutanol, and removing acid components with an aqueous saturated or near-saturated aqueous salt solution, chemically drying the biobutanol with at least one absorbent, and distilling the biobutanol to produce a purified biobutanol product.

[0009] Another aspect of the invention relates to a system that isolates biobutanol including a fermentation vessel that removes biobutanol from a fermentation using a separation system that yields an organic layer of concentrated biobutanol, a dehydrating device that dewaters the concentrated biobutanol and removes acid components with an aqueous saturated or near-saturated aqueous salt solution, a chemical drying device that dries the biobutanol with at least one absorbent, and a distilling system that separates undesired components from the biobutanol to produce a purified biobutanol product.

[0010] Embodiments of the invention further include utilizing at least one inert gas during the separation process. In embodiments, the separation process is selected from the group consisting of a sparging process, membrane separation, and continuous feed by mechanical stirring of said broth. In embodiments, the dewatering includes passing said concentrated biobutanol over a stripper to remove acetone. In embodiments, the salt solution is a brine solution. In embodiments, the distilling system includes a fractionation process. In other embodiments, the distilling process includes a fractionation process having between 1 to 20 theoretical plates, or 10. In embodiments, the aqueous salt solution includes inorganic salts with sodium or potassium ions and negative ions of chloride, sulfate, carbonate, and/or bicarbonate at saturation. The salt solution can have a pH of about 8-10.

[0011] The purified biobutanol product can be about 97-99+% pure 1-butanol. In embodiments, the biobutanol is iso-butanol, using collection method temperatures of about 107-112°C, or 107-110°C. In embodiments, the salt solution is within 80-99% of saturation. In other embodiments, the salt solution is within 80 to 99% of saturation or near saturation, or 95% of saturation. In embodiments, the device that dewaters the biobutanol results in about 1-4% water remaining in the purified biobutanol product. In other embodiments, the dewatering of the biobutanol resulting in about 1-4% water remaining, or 2-3%, in the purified biobutanol product. In another aspect of the invention, a purified biobutanol product is obtained from the process defined above.

[0012] Embodiments of the invention further include utilizing at least one inert gas during the separation system. In embodiments, the separation system is selected from the group consisting of a sparging process, membrane separation, and continuous feed by mechanical stirring of the broth. In embodiments, the dehydrating device includes a stripper to remove acetone.

[0013] Embodiments of the invention can lead to an increase in production yield, simplify the equipment, lower energy use in the purification process, and concurrently eliminating acidic components. The brine solution-drying/purification method can be coupled with other techniques including sparging by carbon dioxide to remove and collect the biobutanol. The latter also requires a complicated distillation that could be mitigated by treatment with the brine solution.

[0014] Typical alternatives could be membrane technology, although this process can suffer from difficulty in running at large scale and continuous operation. As it stands several distillations are required and involve reduced yields from losses in the distillation-only processing. Embodiments of the invention could be employed with other alcohols like pentanol, which is also a target of the fermentation business of making new biofuels.

[0015] An embodiment of the invention includes a method demonstrating a quick and efficient way to remove water and acids from butanol by contacting the organic phase with a near-saturated aqueous salt solution which rapidly dewaters the butanol and removes acid components. This can be followed by chemical drying using a minimum of absorbent and then the product distilled to afford pure butanol with exceptional mass yields and low energy consumption.

[0016] The butanol broth used herein can be prepared by one of several methods known to those skilled in the art of fermentation. Typically, once a maximum titer is achieved in the fermentation vessel, the butanol is removed in some form of a sparging process whereby the butanol is concentrated and then phase separates as an organic layer that can include by-products of the fermentation process that may have organic chemicals/contaminants including, but not limited to, acetone, ethanol, furans, acetic acid, butyric acid, and various esters. Depending on the mixture's exact composition, 15-20 wt. % of water will also remain in the butanol. While 15-20 wt. % is a common amount of the remaining water, water contents from

5 to 40 wt. % are generally acceptable.

[0017] The near-saturated or saturated salt solution used herein can be formed by treating water with an excess amount of an inorganic salt. Typical inorganic salts that can be used include sodium and potassium ions with negative ions of one or more of chloride, sulfate, carbonate, and bicarbonate, and combinations thereof, but the method is not limited to these. One factor for the resulting salt solution is that it be sufficiently close to or at saturation. Once in process, the aqueous salt solutions can be brought to near saturation by evaporation water by an active heating/evaporation process, outdoor evaporation in open ponds, or in a recycling system using evaporation to remove water and concentrate the salt solution. Near saturation generally refers to a salt solution in which at least one inorganic salt is present at concentration sufficient for the salt solution, at the temperature when it is in contact with the broth, to be at 90% or more of the theoretical weight present at full saturation, such as at least 95% or at least 98 wt. % of the theoretical weight.

[0018] The salt solution is brought to a pH of 8-10, such as about 8. The basic salt solution is brought in contact with the butanol sparge feed and mixed thoroughly by mechanical stirring for a period of 5 to 30 min, e.g., 15 min. The top organic phase is the dewatered butanol, with any acid contaminants being removed into the salt solution. The top organic phase is passed over a stripper to remove acetone and then transferred through an optional secondary drying column and then on to a continuous distillation column for final fractionation. The bio-n-butanol can be further dried with exposure to a drying agent. This can bring the water content to below1% (by weight). Collection of butanol from a 113-117 °C fraction affords 97-99+ % pure 1-butanol with capture of 90-99% of the theoretical biobutanol included in the "biobutanol beer broth." Similar results can be obtained for iso-butanol using an identical process except collecting a fraction at 107-110 °C. The bottoms in the final and only distillation can be discarded in the process and typically represent 1-2 wt. % of the volume of purified butanol. They typically include salt residue that can be recycled.

[0019] In **FIG 1**, a general overview of process/apparatus going from sugars (fermentation) to purified biobutanol is shown. Biomass**10** undergoes a fermentation process and at S100 initial removal of biobutanol takes place to provide crude biobutanol **12**. At S102, azeotropic or other

distillation may take place. Other organic components may be phase separated from the biobutanol. The low cut **14** is taken which is an azeotropic mixture of biobutanol and water and may include other products of the fermentation. At S104, this fraction **14** is treated with a brine solution at or near saturation. Additional salt may be added to the contacting brine solution to ensure the brine is as close to saturation as possible. The salt/water **18** and biobutanol layer **18** are allowed to separate and at S106, the top biobutanol layer 18 is taken and may be subjected to further separation methods at S106, such as drying in a drying column and a final distillation to yield biobutanol **22**. The brine solution **16** may be treated at S108to recover the salt for reuse.

[0020] **FIG 2** shows a process flow diagram (PFD) and apparatus for dewatering and contaminant removal from "biobutanol beer broth" obtained as a sparge from a fermentation vessel **30**. The sparge feed is distilled in an azeotropic distillation apparatus **30** and the azeotropic biobutanol feed is treated with salt in a tank **32** and the salt solution is fed to a concentrator (e.g., evaporator) **34** where excess water is removed by evaporation and/or additional salt is added to maintain the salt solution at or near saturation. The concentrator **34** is configured for regulating the pH of the aqueous salt solution to maintain a predetermined pH (8-10) or salt concentration (within a predetermined range of saturation (e.g., 95 wt. % -100%) in the aqueous salt solution within the dewatering device **34**.

[0021] The saturated solution is then returned to the tank **32**. The salt solution concentration can be a continuous or semi-continuous process. The biobutanol feed is fed to a stripper **36**, where acetone is removed from the feed. The treated biobutanol feed is passed through a drying column **38** and the further dried biobutanol feed is fed to a distillation column **40**, where a butanol-rich fraction is removed which can be at least 97 wt. %, or at least 98 wt. %, or at least 99 wt. % butanol.

[0022] **FIG 3**. Proton nuclear magnetic resonance (NMR) spectrum of dewatered bio-1-butanol obtained from an original "butanol beer broth" including 15-18 wt. % water. The original broth was obtained by fermentation of sugars using Clostridium bacteria.

Example

[0023] At S100, bio-n-butanol is removed from the fermentation vessel **28** by a process of a) sparging with an inert gas, b) membrane separation, and/or c) some other physical means that

will remove the bacteria, yeast, or fungi and remove water and concentrate the biobutanol component.

[0024] At S102, the bio-n-butanol azeotrope (~55% butanol, 45% water, b.p. 97 °C) is carefully distilled using an apparatus **30** with sufficient theoretical separation plates to minimize additional water (bp 100 deg C) co-distilling with the azeotrope. In addition to the bio-n-butanol azeotrope other organics and azeotropes will lead the distillation run. They can be discarded or left in the distillate and will be removed in subsequent steps.

Version A

[0025] At S104, after settling, the azeotrope will form two layers, sodium chloride is added at a sufficient amount to saturate the water layer, the layers are separated and the butanol is isolated by a quick physical separation and can be used in certain processes, for example, dehydration to form bio-1-butene.

Version B

[0026] At S104, the water layer can be separated (discarded or used in a slower secondary process to remove the butanol) from the biobutanol, leaving a water saturated biobutanol. This biobutanol is treated with a saturated salt solution, typically in the range of 10-50% of the volume of the butanol volume, thus removing most of the water. The layers are separated by quick physical separation and the brine solution can be strengthened (i.e. salt added and/or water evaporated off) and used many times.

[0027] At S106, an optional secondary process to remove additional water can be utilized. This can be 3A mol-sieves or a related solid-phase drying agent that can be recycled in service by heating or similar. **Fig. 3** shows a typical dried bio-n-butanol sample using this process.

[0028] Depending on the biobutanol specs, a final distillation to remove last traces of water, ethanol, acetone, and other minor impurities may be desirable. For many applications, this biobutanol may be of sufficient purity, for example use in preparing biobutene that is useful in making diesel and jet fuels and biolubricants.

[0029] Where a range of values is provided, it is understood that each intervening value, to the tenth of the unit of the lower limit unless the context clearly dictates otherwise, between the upper and lower limits of that range is also specifically disclosed. Each smaller range between

any stated value or intervening value in a stated range and any other stated or intervening value in that stated range is encompassed within the invention. The upper and lower limits of these smaller ranges may independently be included or excluded in the range, and each range where either, neither or both limits are included in the smaller ranges is also encompassed within the invention, subject to any specifically excluded limit in the stated range. Where the stated range includes one or both of the limits, ranges excluding either or both of those included limits are also included in the invention.

CLAIMS

What is Claimed is:

1. A purification process of isolating biobutanol, comprising:

removing biobutanol from a fermentation broth using at least one separation process to phase separate a concentrated organic layer which includes biobutanol;

dewatering said concentrated biobutanol and removing acid components with a saturated or near-saturated aqueous salt solution;

chemically drying said biobutanol with at least one absorbent; and

distilling said biobutanol to produce a purified biobutanol product.

2. The process according to claim 1, further characterized by utilizing at least one inert gas during the separation process.

3. The process according to claim 1, further characterized by said separation process being selected from the group consisting of a sparging process, membrane separation, and continuous feed by mechanical stirring of said broth.

4. The process according to claim 1, further characterized by said dewatering includes passing said concentrated biobutanol over a stripper **36** to remove acetone.

5. The process according to claim 1, further characterized by said aqueous salt solution including at least one inorganic salt of sodium or potassium ions with negative ions of at least one of chloride, sulfate, carbonate, and bicarbonate.

6. The process according to claim 1, further characterized by said salt solution being a brine solution.

7. The process according to claim 1, further characterized by said salt solution being at

saturation.

8. The process according to claim 1, further characterized by said salt solution including at least one of sodium or potassium cations with negative ions of at least one of chloride, sulfate, carbonate, and biocarbonate and which is maintained within 80 to 99% of saturation, or within 95% of saturation.

9. The process according to claim 1, further characterized by regulating the pH of the salt solution to have a pH of 8-10.

10. The process of claim 1, further characterized by the dewatering including recycling the salt solution through a concentrator device **34** which increases a concentration of salt in the salt solution and returning the more concentrated salt solution for dewatering the concentrated biobutanol.

11. The process according to claim 1, further characterized by said distilling process including a fractionation process having between 1 to 20 theoretical plates, or 10.

12. The process according to claim 1, further characterized by said purified biobutanol product being at least 97% pure 1-butanol, or at least 98% or at least 99% pure 1-butanol.

13. The process according to claim 1, further characterized by said purified biobutanol product being at least 97% pure 2-butanol, or at least 98% or at least 99% pure 2-butanol.

14. The process according to claim 1, further characterized by said biobutanol being iso-butanol and the distilling includes collecting a fraction at a temperature of 107-112°C.

15. The process according to claim 1, further characterized by said biobutanol being 1-butanol and the distilling distilling includes collecting a fraction at a temperature of 113-117 °C.

16. The process according to claim 1, further characterized by said dewatering of said biobutanol resulting in up to 4% water remaining in said purified biobutanol product.

17. The process according to claim 16, further characterized by said dewatering of said biobutanol resulting in 1-4% water remaining, or 2-3% water remaining, in said purified biobutanol product.

18. The process according to claim 1, further characterized by said dewatering being used for continuous extraction from a feed of the concentrated biobutanol.

19. A purified biobutanol product obtained from the process defined in any one of claims 1-18.

20. A system that isolates biobutanol, comprising:

a fermentation vessel **28** that removes biobutanol from a fermentation using a separation system **30** that yields an organic layer **14** of concentrated biobutanol;

a dehydrating device **32** that dewaters a said concentrated biobutanol and removes acid components with a saturated or near-saturated aqueous salt solution;

a chemical drying device **38** that dries said biobutanol with at least one absorbent; and

a distilling system **40** that separates undesired components from said biobutanol to produce a purified biobutanol product.

21. The system according to claim 20, further characterized by utilizing at least one inert gas during the separation system.

22. The system according to claim 20, further characterized by said separation system being selected from the group consisting of a sparging process, membrane separation, and continuous feed by mechanical stirring of said broth.

23. The system according to claim 20, further characterized by said dehydrating device including a stripper to remove acetone.

24. The system according to claim 20, further characterized by said salt solution being a brine solution.

25. The system according to claim 20, further characterized by said distilling system including a fractionation system.

26. The system according to claim 20, further characterized by said salt solution including an inorganic salt solution containing sodium or potassium ions with negative ions of at least one of chloride, sulfate, carbonate, and bicarbonate.

27. The system according to claim 20, further characterized by said salt solution having a pH of 8-10.

28. The system according to claim 20, further characterized by said purified biobutanol product being at least 97% pure 1-butanol.

29. The system according to claim 20, further characterized by said biobutanol being iso-butanol and said distilling system configured for collection of a fraction at a temperature of 107-112°C.

30. The system according to claim 20, further characterized by a concentrator **34** which maintain said salt solution being within 80-99% saturation, by at least one of evaporating water and adding salt to provide sodium or potassium ions with negative ions of at least one of chloride, sulfate, carbonate, and biocarbonate.

31. The system according to claim 20, further characterized by said dehydrating device **32**

dewatering said biobutanol resulting in 1-4% water remaining in said purified biobutanol product.

32. The system according to claim 20, further characterized by the dehydrating device **32** being configured for extraction from a continuous feed of the concentrated biobutanol.

32. The system according to claim 30, further characterized by the concentrator device **34** being configured for regulating the pH of the aqueous salt solution to maintain at least one of a predetermined pH and a predetermined salt concentration in the aqueous salt solution within the dewatering device **32**.

33. The system according to claim 30, further characterized by the predetermined pH being 8-10.

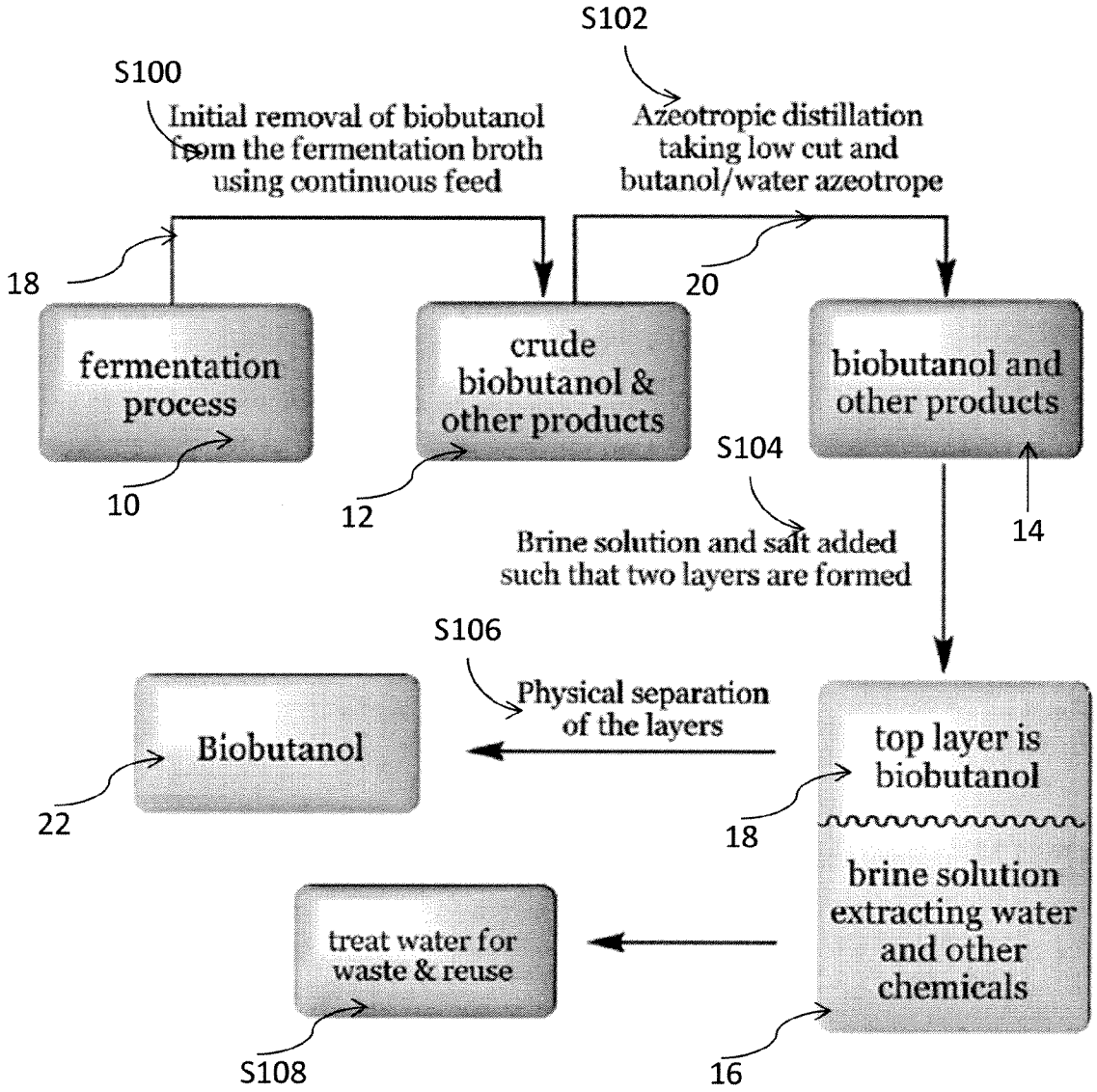

S102
S100
Initial removal of biobutanol from the fermentation broth using continuous feed
Azeotropic distillation taking low cut and butanol/water azeotrope
18
20
fermentation process
crude biobutanol & other products
biobutanol and other products
10
12
S104
14
Brine solution and salt added such that two layers are formed
S106
Physical separation of the layers
Biobutanol
top layer is biobutanol
22
18
brine solution extracting water and other chemicals
treat water for waste & reuse
S108
16

FIG. 1

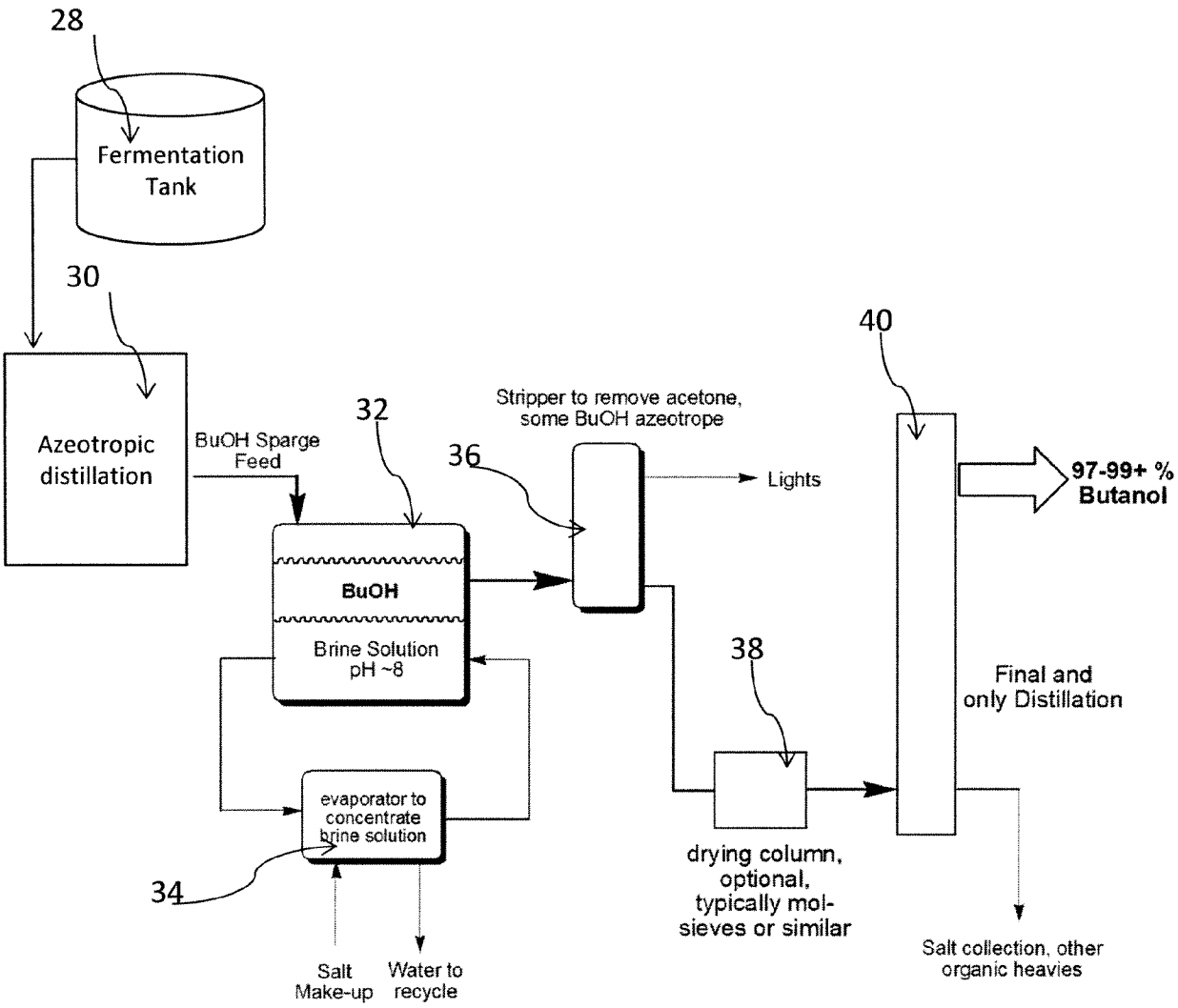

28
Fermentation Tank
30
Azeotropic distillation
BuOH Sparge Feed
32
BuOH
Brine Solution pH ~8
34
evaporator to concentrate brine solution
Salt Make-up
Water to recycle
36
Stripper to remove acetone, some BuOH azeotrope
Lights
38
drying column, optional, typically mol-sieves or similar
40
97-99+ % Butanol
Final and only Distillation
Salt collection, other organic heavies

FIG. 2

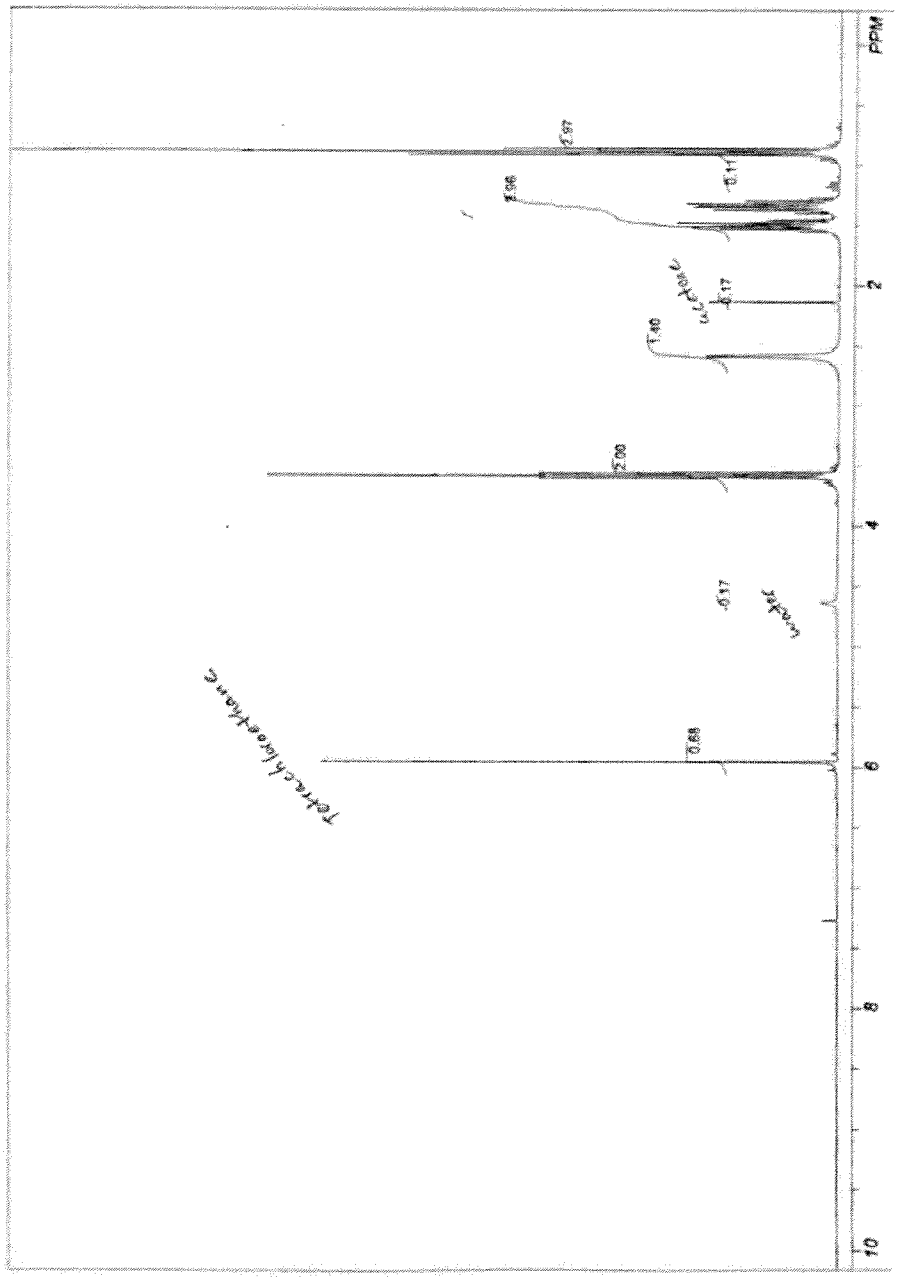

PPM
2
4
6
8
10
2.97
0.11
3.06
1.17
1.80
2.00
0.17
0.88
Tetrachloroethane

FIG. 3

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US 12/35132

A. CLASSIFICATION OF SUBJECT MATTER
IPC(8) - C12P 7/16, B01D 3/00, C02F 1/04 (2012.01)
USPC - 435/160, 210/633, 210/600
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
USPC -- 435/160, 210/633, 210/600, 435/157, 435/155, 435/132, 435/41, 210/664, 210/188

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
PubWEST -- PGPB,USPT,USOC,EPAB,JPAB; Dilaog Classic Files -- 654, 652, 349, 348, 340, 35, 65, 155; USPTO Web Page; Google Scholar; Search terms -- purification, 1-butanol, 2-butanol, fermentation broth, saturated salt, phase separation, distillation, fractionation, apparatus, dewatering, sodium chloride, brine, inert gas, sparging, membrane separat

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 2008/0132730 A1 (MANZER et al.) 05 June 2008 (05.06.2008) para [0007], [0010], [0016], [0042]-[0049], [0051], [0053], [058], [0068]-[0071], Fig 2-5	1-31, 32A, 32B, 33
Y	US 2011/0111475 A1 (KUHRY et al.) 12 May 2011 (12.05.2011) para [0013], [0019], [0045], [0048], [0057], [0087]	1-31, 32A, 32B, 33
Y	US 2010/0155333 A1 (HUSAIN et al.) 24 June 2010 (24.06.2010) para [0004], [0023], [0025], [0036]-[0038], [0046], [0048]	1-31, 32A, 32B, 33
Y	US 2008/0102502 A1 (FOODY et al.) 01 May 2008 (01.05.2008) para [0042], [0045], [0049], [0055]-[0060], [0118], [0119]	10, 19/10, 30, 32B
Y	US 4,473,444 A (FELDMAN et al.) 25 September 1984 (25.09.1984) col 4, ln 51-55; col 5, ln 34-56, Table 1	11, 14, 15, 19/(11,14,15), 25, 29
Y	US 2002/0177728 A1 (BOUDREAUX et al.) 28 November 2002 (28.11.2002) para [0051], [0053]	11, 19/11

☐ Further documents are listed in the continuation of Box C. ☐

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" earlier application or patent but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
27 June 2012 (27.06.2012)	24 JUL 2012
Name and mailing address of the ISA/US Mail Stop PCT, Attn: ISA/US, Commissioner for Patents P.O. Box 1450, Alexandria, Virginia 22313-1450 Facsimile No. 571-273-3201	Authorized officer: Lee W. Young PCT Helpdesk: 571-272-4300 PCT OSP: 571-272-7774

Form PCT/ISA/210 (second sheet) (July 2009)

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
18 July 2013 (18.07.2013)

WIPO | PCT

(10) International Publication Number
WO 2013/106065 A1

(51) **International Patent Classification**:
B01J 31/00 (2006.01) ***B01J 37/00*** (2006.01)

(21) **International Application Number**:
PCT/US2012/035167

(22) **International Filing Date**:
26 April 2012 (26.04.2012)

(25) **Filing Language**: English

(26) **Publication Language**: English

(30) **Priority Data**:

61/585,943	12 January 2012 (12.01.2012)	US
13/426,118	21 March 2012 (21.03.2012)	US
13/426,192	21 March 2012 (21.03.2012)	US

(71) **Applicant** *(for all designated States except US)*: **THE GOVERNMENT OF THE UNITED STATES OF AMERICA AS REPRESENTED BY THE SECRETARY OF THE NAVY** [US/US]; Naval Air Warfare Center - Weapons Division, 1 Administration Circle, Stop 1009, China Lake, CA 93555-6100 (US).

(72) **Inventor; and**
(75) **Inventor/Applicant** *(for US only)*: **WRIGHT, Michael, E.** [US/US]; 245 N. Alvord, Ridgecrest, CA 93555 (US).

(74) **Agents**: **HALEY, Charlene A.** et al.; Naval Air Warfare Center, Weapons Division, 1 Administration Circle, China Lake, CA 93555-6100 (US).

(81) **Designated States** *(unless otherwise indicated, for every kind of national protection available)*: AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, JP, KE, KG, KM, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) **Designated States** *(unless otherwise indicated, for every kind of regional protection available)*: ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Published:

— *with international search report (Art. 21(3))*

(54) **Title**: HOMOGENEOUS METALLOCENE ZIEGLER-NATTA CATALYSTS FOR THE OLIGOMERIZATION OF OLEFINS IN ALIPHATIC-HYDROCARBON SOLVENTS

(57) **Abstract**: A process is disclosed that permits continuous batch conversion of alpha-olefins and internal-olefins to oligomeric materials without fouling the reaction vessel and provides a simple and highly efficient method for making very cost effective catalyst systems based on Zeigler-Natta Group 4 metallocenes. Embodiments of this invention produce diesel and turbine fuels that are 100% synthetic iso-paraffinic kerosenes with flashpoints greater than 61 °C.

WO 2013/106065 A1

HOMOGENEOUS METALLOCENE ZIEGLER-NATTA CATALYSTS FOR THE OLIGOMERIZATION OF OLEFINS IN ALIPHATIC-HYDROCARBON SOLVENTS

STATEMENT REGARDING FEDERALLY SPONSORED RESEARCH OR DEVELOPMENT

[0001] The invention described herein may be manufactured and used by or for the government of the United States of America for governmental purposes without the payment of any royalties thereon or therefor.

FIELD OF THE INVENTION

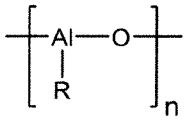
[0002] The invention generally relates to an approach that permits continuous batch conversion of alpha-olefins and internal olefins to oligomeric materials without fouling the reaction vessel and provides a simple and highly efficient method for making very cost effective catalyst systems based on Group 4 metallocenes. Embodiments of the invention create synthetic iso-paraffinic kersosenes with high flashpoints suitable for use gas, diesel, and turbine engines.

SUMMARY

[0003] A process for preparing an Activated Homogeneous Metallocene Catalyst (AHMC) includes contacting a metallocene precatalyst of the formula (I):

$$L^1L^2MX^1X^2 \qquad (I)$$

with an alkylaluminoxane of formula (II):



(II);

[0004] The precatalyst, alkylaluminoxane and/or their combination is combined with at least one saturated hydrocarbon solvent. M is Ti, Zr, or Hf. X^1 and X^2 are independently at least one of

halogen, hydrogen, alkyl, and a mixture thereof. L^1 and L^2 are independently at least one π-ligand selected from the group consisting of cyclopentadienyl, indenyl, fluorenyl, and derivatives thereof. n is an integer of at least 3. R is a C_1-C_{30} alkyl. The at least one saturated hydrocarbon solvent can be selected from the group consisting of cycloalkanes, normal alkanes, iso-alkanes, and combinations thereof.

[0005] In another aspect, a process for oligomerizing an α-olefin can be performed by contacting an olefin feedstock with the Activated Homogeneous Metallocene Catalyst to form an olefinic-oligomer product, the olefin feedstock including at least one olefin, such as an α-olefin.

BRIEF DESCRIPTION OF THE DRAWINGS

[0006] **FIG. 1** is a photograph of a beaker removed directly from SS-bomb showing a typical homogeneous oligomeric mixture from 1-butene that contains the Cp_2ZrCl_2/MAO catalyst system prepared in cyclohexane, according to embodiments of the invention.

[0007] It is to be understood that the foregoing general description and the following detailed description are exemplary and explanatory only and are not to be viewed as being restrictive of the invention, as claimed. Further advantages of this invention will be apparent after a review of the following detailed description of the disclosed embodiments, which are illustrated schematically in the accompanying drawings and in the appended claims.

DETAILED DESCRIPTION OF EMBODIMENTS OFTHE INVENTION

[0008] The invention generally relates to an approach that permits continuous batch conversion of alpha-olefin and internal-olefins to oligomeric materials without fouling the reaction vessel and provides a simple and highly efficient method for making very cost effective catalyst systems based on Group 4 metallocenes.

[0009] Currently no homogeneous Ziegler Natta (ZN) catalyst can be used in the absence of aromatic solvents. Aspects of the exemplary embodiments disclosed herein provide a versatile and inexpensive means for creating and using highly active catalysts for the oligomerization of alpha-olefins, like 1-butene. Hence, the method is very valuable for turning 1-butene (made from bio-1-butanol) to oligomers suitable for use in creating diesel and jet fuels with flashpoints over 61 deg C.

Furthermore, by addition of a homogeneous isomerization catalyst mixtures of alpha-olefins and internal-olefins can be converted to high flashpoint diesel and jet fuels using a homogeneous ZN catalyst prepared as disclosed herein. The exemplary activated catalyst can be used in oligomerization and/or dimerization of olefins in the absence of any aromatic solvents (i.e., where aromatic solvents make up less than 5 wt.% or less than 1 wt. % of the liquid reaction mixture).

[0010] In various aspects, an activated catalyst, a method for making it and a method for using it to oligomerize/dimerize an olefin are disclosed.

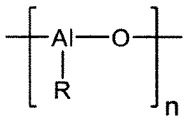
[0011] One aspect of the invention includes a fuel(s). The fuel(s) can be produced from oligomerization and/or dimerization processes including at least one alpha-olefin and/or internal olefins, at least one homogenous Ziegler-Natta catalyst and at least one homogenous activating co-catalyst (e.g., together forming the exemplary activated catalyst), and at least one or mixture hydrocarbon solvent, where the fuel has a flashpoint of 61 to 100 °C. Embodiments of the invention further include at least one olefin-isomerization catalyst. Other embodiments of the invention include the Ziegler-Natta catalyst being group four (4) metallocene catalyst. In other embodiments, the co-catalyst is an alkylaluminoxane (AAO). Yet in other embodiments, the catalyst is prepared by contacting a metallocene precatalyst with an aliphatic hydrocarbon solution containing an alkylaluminoxane (AAO). In still yet other embodiments, the aliphatic hydrocarbon solution is derived from a fuel.

[0012] Embodiments of the invention include the activated catalyst being prepared by dissolving trialkylalane in an aliphatic-hydrocarbon solvent (e.g., a fuel) and being treated with one mol-equivalent of water and then contacted with a metallocene precatalyst and filtered. In embodiments, the trialkylalanes are selected from the group consisting of trimethylalane, triethylalane, tributylalane, and tri(iso-butyl)alane (TIBA). In other embodiments, the aliphatic hydrocarbon solvents (which can be fuels) are selected from the group consisting of straight chain alkanes including hexane, heptane, octane, nonane, decane, and alkanes having greater than 10 carbons. In yet other embodiments, the aliphatic hydrocarbon solvents are C6 to C16 hydrocarbons and may be selected from the group consisting of, but not limited to, branched-aliphatic-hydrocarbons including 3-methylheptane, 2-methyloctane, and the like. Still yet in other embodiments, the aliphatic hydrocarbon solvents are selected from the group consisting of, but not limited to, cyclic aliphatic-hydrocarbons including cyclohexanes, methylcyclohexanes,

dimethylcyclohexanes, tetralins, pinanes, and other mono- and bicyclic aliphatic-hydrocarbons.

[0013] Another aspect of the invention includes a process for preparing a saturated hydrocarbon fuel including contacting an olefin feedstock with an Activated Homogeneous Metallocene Catalyst (AHMC) for use in the dimerization and oligomerization of olefins, such as neat α-olefins, where the AHMC can be prepared by contacting a metallocene precatalyst of the formula (I) with an alkylaluminoxane of formula (II), where the latter is prepared in and/or the resulting activated catalyst mixed with a saturated hydrocarbon solvent:

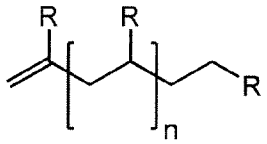
$$L^1L^2MX^1X^2 \qquad (I)$$



(II)

[0014] In formula ((I), M is Ti, Zr, or Hf, X^1 and X^2 is at least one halogen, hydrogen, alkyl, or a mixture thereof, L^1 and L^2 are comprised of π-ligands selected from the group consisting of cyclopentadienyl, indenyl, fluorenyl and derivatives thereof. In the derivatives, substituents are attached to the ligands, the attachments includes ansa-linkages that bind the two rings, with at least one carbon or silicon atoms each bearing hydrogen or alkyl radicals. In formula (II), n is an integer of at least 3, and R can be a C_1-C_{30} alkyl, which can be linear, branched, or a combination thereof. The at least one saturated hydrocarbon solvent can be selected from the group consisting of cycloalkanes, normal-alkanes, and/or iso-alkanes.

[0015] The Al/M molar ratio can be about 1 to 100. In other embodiments, the Al/M molar ratio is 10 to 20. Embodiments of the invention include the product being prepared by placing the activated catalyst in contact with a neat solution having at least one α-olefin during a period of 1 to 24 h and forms the α-olefinic-oligomer (III) in 98-100% conversion based on starting α-olefins:



(III)

[0016] In formula (III) each R can be independently as described for formula (II).

[0017] Embodiments of the invention include the AHMC being introduced in one portion, in discrete aliquots, or through a continual addition process directly to the neat (undiluted) α-olefin

feedstock. Embodiments of the invention include the ratio of metallocene precatalyst (g of M) to α-olefinic-oligomer product (Kg) being about 5 to 40. In other embodiments, the entire α-olefinic-oligomer product (III) is transferred to quench vessel, treated with water, and then filtered to remove solids. In other embodiments, the α-olefinic-oligomer product (III) is passed through an alumina or other metal oxide filter material to remove M and Al species from the product solution. Other embodiments, the α-olefinic-oligomer product (III) is treated with a stabilizing agent selected from either a hydroquinone or phenol family of stabilizers, e.g., with butylated hydroxytoluene (BHT) and at concentrations of 10-300 ppm.

[0018] Embodiments of the invention include the process lowering the boiling fraction of the α-olefinic-oligomer products (III) between 50 °C and 150 °C, including between 70 °C and 130 °C, and are removed by flash distillation. Embodiments of the invention include the process having a low boiling fraction from about 25 to 45 wt. % of the total α-olefinic-oligomer product, or comprises about 30-40 wt. % of the total α-olefinic-oligomer. In embodiments, the process includes a light boiling fraction subjected to selective-dimerization chemistries to create a mixture of mono-olefinic hydrocarbon products having a boiling point greater than 200 °C but less than 300 °C at atmospheric pressure. Embodiments of the invention include the process having a high boiling fraction subjected to catalytic hydrogenation over heterogeneous metal catalysts having palladium, platinum, and/or nickel (or any combination thereof). Embodiments of the invention include the process having a hydrogen pressure in the range of about 30-3000 psig (approximately 300-20800 kpa), or a pressure between 50 and 100 psig (approximately 440-790 kpa). Embodiments of the invention the reaction temperature used is in the range of about 20 to 100 °C with ambient or 40 °C temperature range.

[0019] Embodiments of the invention further include the product being subjected to hydrogenation over heterogeneous metal catalysts which includes palladium, platinum, and/or nickel; hydrogen pressure is performed in the range of about 30-3000 psig, or a pressure between 1000 and 2000 psig. In embodiments, the temperature is in the range of 20 to 100 °C or 60 °C to 80 °C temperature range. In embodiments, the products are combined and distilled at atmospheric pressure. In other embodiments, the products obtained are combined and distilled at reduced pressure. Embodiments of the invention a collection of hydrocarbon, a completely 100% synthetic iso-paraffinic kerosene (SiPK) product, starting at 150 °C and finishing at 280 °C affords a fuel

mixture that meets Jet-A/JP-8 and diesel #1 flashpoint requirements (≥38 °C). Embodiments of the invention the SiPK jet/diesel fuel produced has a derived Cetane index of 40-55 or 50 Cetane index. In other embodiments, the SiPK jet/diesl fuel produced has a maximum cold flow viscosity of 8 cSt as measured at -20 °C (ASTM 445LT).

[0020] Embodiments of the invention include the collection of product starting at 170 °C and finishing at 280 °C yields a SiPK jet/diesel that meets military JP-5 and diesel #2 flashpoint requirements with a flashpoint >61 °C. Embodiments of the invention include the SiPK jet/diesel fuel produced has a derived Cetane index of 40-55 or 50 Cetane index. Embodiments of the invention include SiPK jet/diesl fuel produced has a maximum cold flow viscosity of 8.5 cSt as measured at -20 °C (ASTM 445LT).

[0021] The α-olefins can be selected from the group consisting of, but not limited to, 1-propene, 1-butene, 1-pentene, and 1-hexene, with neat 1-butene or a mixture of 1-butene and 1-propene (3:1, mol:mol, respectively).

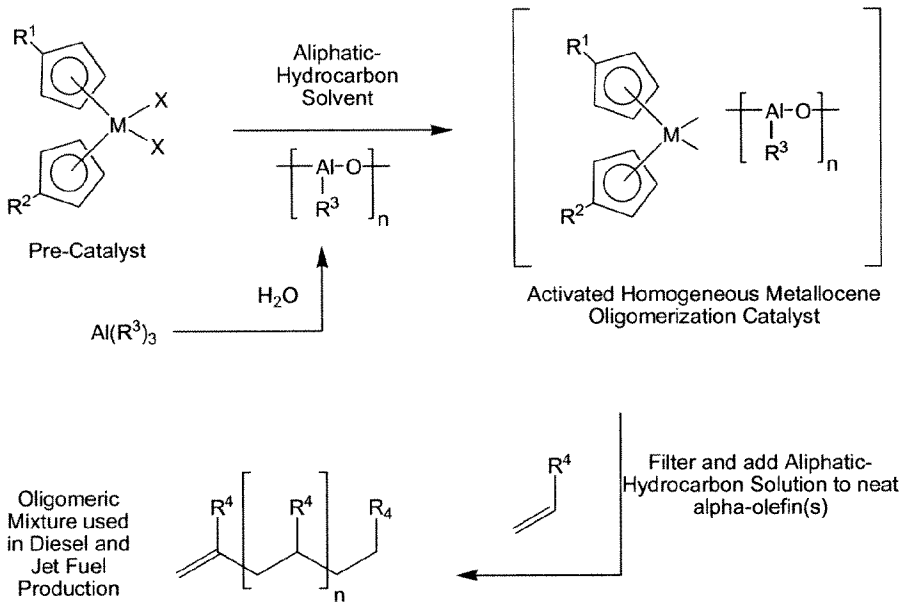
[0022] In embodiments of the invention the π-ligands can be cyclopentadienyl, R is methyl, and the aliphatic hydrocarbon solvent is cyclohexane or methylcyclohexane. Embodiments of the inventions include the π-ligands are cyclopentadienyl rings having at least one alkyl group attached, R is methyl, and the aliphatic hydrocarbon solvent is selected from the group consisting of cyclic, acyclic linear and branched hydrocarbon solvent that include jet and diesel fuels prepared. Embodiments of the invention include the π-ligands being cyclopentadienyl, R is at least three carbons, and the aliphatic hydrocarbon solvent is selected from acyclic linear and/or branched hydrocarbon solvent.

[0023] Embodiments of the invention include the trialkylalane compound being prepared by direct reaction of one mole of trialkylalane with one mole-equivalent of water in an SiPK. Other embodiments of the invention include the trialkylalane is selected from the group consisting of, but not limited to, trimethyl, triethylalane, tripropylalane, tri-isopropylalane, tributylalane, and tri-isobutylalane, and tri-isobutylalane. Still yet in other embodiments the π-ligands are cyclopentadienyl rings having at least two alkyl groups attached, R is methyl, and the aliphatic hydrocarbon solvent is selected from the group consisting of cyclic, acyclic linear, branched hydrocarbon solvents, solutions thereof, and jet and diesel fuels previously prepared. Embodiments of the invention include a fuel having a flashpoint of about 61 to about 100. Embodiments of the

invention further include an isomerization catalyst having a range from about 0.1 ppm to about 0.1 weight %, relative to the total olefin component.

[0024] Oligomers, suitable for conversion and use as jet and diesel fuels, are prepared from alpha-olefins using highly active purely homogeneous Ziegler-Natta catalysts, a homogeneous co-catalyst, and in pure aliphatic hydrocarbon solvent. The oligomerization can be conducted controlled without the use of hydrogen and produce an ideal molecular distribution of oligomers for use in preparing diesel and jet fuels. This method creates fuels that do not require any subsequent hydrocracking or reforming yet retain outstanding cold flow properties and exceptionally high gravimetric densities.

[0025] The homogeneous catalyst of embodiments of the invention is prepared as shown in Scheme 1 by contacting a metallocene precatalyst with a solution of an alkylaluminoxane (AAO). Typically, R^1 and R^2 can independently be hydrogen or any combination of C_1-C_{30} alkyl chains. The R^1 and R^2 substituents(s) are linear or branched chains and/or a combination thereof. The alkylaluminoxane can be prepared using methods that are familiar to people skilled in the art, e.g., from a trialkylalane $Al(R_3)_3$ where R^3 is typically a C_1-C_{30} alkyl group, although larger chains can be employed. Branching in the alkyl group can be used to increase solubility although is not required herein. When the AAO is prepared in an aromatic solvent, the metallocene precatalyst is combined and the aromatic solvent is removed under reduced pressure and the solids redissolved in the desired aliphatic-hydrocarbon solvent or fuel, filtered if necessary, and then transferred to the reaction vessel that contains the mixture of alpha-olefin and internal-olefins. These activated homogeneous catalyst solutions are stable and suitable for storage. Typical alpha-olefins can include R^4 being a C_1-C_{10} alkyl group that can be a linear chain or contain branching at various places in the carbon chain. The internal olefin is any olefin-isomer where the double-bond has chemically migrated internally along the carbon chain. The homogeneous isomerization catalyst takes the internal-olefin and isomerizes the bond so as to create the corresponding alpha-olefin.

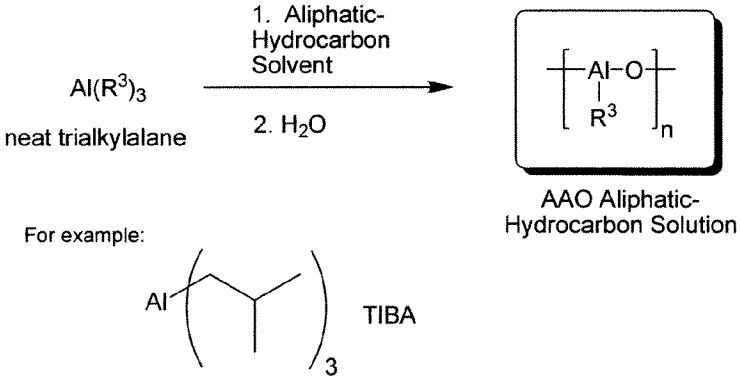


Scheme 1

[0026] Another embodiment of the chemical process for preparing the homogeneous metallocene catalyst in pure aliphatic-hydrocarbon solvent is illustrated in Scheme 2: A trialkylalane is dissolved in an aliphatic-hydrocarbon solvent and treated with one mol-equivalent of water, this solution is then placed in contact with a metallocene precatalyst. The molar ratio of alane/metallocene can be varied from 1:1 to 100:1, respectively, depending on the desired catalytic process and outcome. The final solutions may be filtered to remove any impurities or undesired products resulting from the controlled hydrolysis of the alane and subsequent reaction with the metallocene pre-catalyst. The final solutions are typically light yellow in color and crystal clear to the eye. Typical concentrations of the active homogeneous catalysts are in the range of 0.1M to 0.01M, with lower concentrations possible with equal catalytic activity. These latter solutions can be stored for long periods or immediately transferred to reaction vessels using common techniques that include Teflon® diaphragm pumps or by simple polypropylene syringes, all with equal success and retention of catalyst activity.

[0027] Typical trialkylalanes can be trimethylalane, triethylalane, tripropylalane, tributylalane, and tri(isobutyl)alane (TIBA), which are all commercially available. Alanes containing longer alkyl

chains can be prepared by existing methods familiar to those in the art by simple reaction of an alkyl Grignard with aluminum trichloride. For the methylaluminoxane, toluene solutions can be utilized from commercial sources to prepare the active metallocene catalyst followed by removal and recycling of the toluene. Then the active metallocene/MAO catalyst can be dissolved in the desired aliphatic-hydrocarbon solvent. Use of MAO in this invention it is found preferable to utilize a metallocene precatalyst that has alkyl substitution on one or more of the cyclopentadienyl rings, although this is not required if certain aliphatic hydrocarbon solvents are used. In general, alkyl-substitution of the metallocene rings provides enhanced solubility over a range of aliphatic-hydrocarbons and can be used to increase the activated metallocene catalyst concentration when desired.



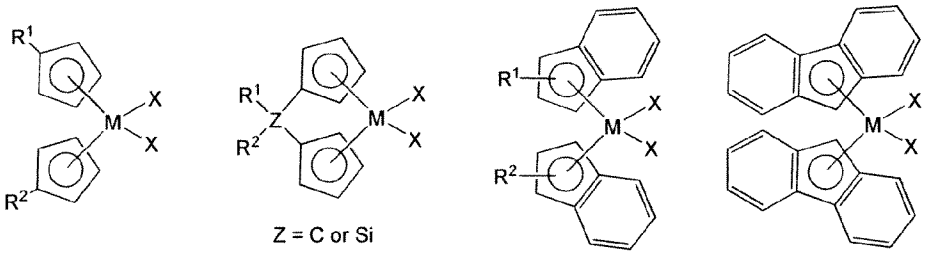
Scheme 2

[0028] Typical aliphatic-hydrocarbon solvents useful in this invention include straight chain alkanes such as hexane, heptanes, octane, nonane, and decane. Branched-aliphatic-hydrocarbons such as 3-methylheptane, 2-methylheptane, and 2-methyloctane work well in this invention as well as other branched aliphatic hydrocarbons containing from 6 to 16 carbons. In particular, C12 to C16 staturated synthetic iso-paraffinic kerosenes are suitable for use in this embodiment. Cyclic aliphatic-hydrocarbons can be used that include cyclohexane, methylcyclohexane, dimethylcyclohexanes, tetralin, pinane, and other mono- and bicyclic aliphatic-hydrocarbons.

[0029] Useful metallocene pre-catalysts for this invention can be prepared by several methods, some found in the open literature covering nearly four decades of group metallocene synthetic

procedures [Y. Qian et al., *Chem. Rev.*, 103(7), pp 2633-2690 (2003); R. Halterman, *Chem. Rev.*, 92(5), pp 965-994 (1992)]. In addition, two patents describe the synthesis of simple bis(n-alkylcyclopentadienyl)MCl_2 (where M = Group 4 transition metal) [J.-S. Oh et al. US6214953 B1 (2001) and J. M. Sullivan et al. US6175027 B1 (2001)]. Oh and co-workers describe the use of alkyl-metallocenes for preparing supported ZN-catalysts and Sullivan and coworkers do not report any catalytic chemistry or solubility properties for the metallocene compounds prepared.

[0030] By way of example, unsubstituted and all alkyl and multiple-alkyl substituted metallocenes, ansa-metallocenes, indenyl, fluorocenyl, and related metallocenes can be used in the present invention when combined with the appropriate aliphatic-hydrocarbon solvent and AAO, all affording highly active and selective oligomerization ZN-catalysts. Examples of the precatalyst (I) include formulas (IV)-(VII) below.



(IV) (V) (VI0) (VII)

[0031] Olefinic monomers that can be used in the current invention include α-olefins (including ethane), mono- and bicyclic olefins, dienes, vinyl-aromatics and the like. Examples include ethene, 1-propene, 1-butene, 1-pentene, 1-hexene, 1-heptene, 1-octene, cyclopentene, cyclopentadiene, norbornene, styrene, and the like. The olefin monomer can be copolymerized with one or more other monomers. Internal-olefins including 2-hexene, 2-pentene, 2-butene can be used in the invention by introduction of a homogeneous group 8 transition metal complex/isomerization catalyst, with nickel being another embodiment of a transition metal.

EXAMPLE 1: Preparation of the Homogeneous Zn-Catalyst in an Aliphatic-Hydrocarbon Solvent

[0032] A Schlenk flask is charged with Cp_2ZrCl_2 (0.080 g, 0.274 mmol) and 10 mL of a toluene MAO solution (8.0 wt. % active MAO). Cp=cyclopentadiene. The toluene was removed

under reduced pressure to afford a yellow solid. To this mixture was added cyclohexane (15 mL) and heated briefly with stirring to dissolve the activated catalyst. The solution was allowed to settle and clear yellow solution was collected by syringe (~10 mL) and used directly or stored for future use.

EXAMPLE 2: Oligomerization of an Alpha-Olefin Using a Homogeneous ZN-Catalyst Delivered in an Aliphatic Hydrocarbon-Solvent

[0033] 1-Butene (~390 g, ~ 500 mL, CP grade) was condensed over calcium hydride inside a Schlenk flask maintained at -70 °C and re-evaporated and passed through a column of activated alumina in route to being condensed inside a dried 0.7L capacity PARR SS bomb. A 500 mL beaker was placed inside the bomb to simulate a glass lined reaction vessel. Once the 1-butene was condensed inside the SS bomb the sample was subjected to five evacuation/backfill cycles to remove residual amounts of oxygen. The homogeneous aliphatic-hydrocarbon solution containing the active catalyst was added to the mixture and 3 addition evacuation/backfill cycles were performed. The SS bomb was sealed and allowed to react for 8 h. Shorter times could be used if the SS bomb was warmed by external heat. Typically after ~1 h the bomb reached a maximum pressure of 80 psig and approached 60 °C in temperature. At completion of the reaction little or no pressure remained in the vessel and the when removed from the SS bomb the beaker contained a clear light yellow solution of oligomers (see Figure 1). Quenching the mixture with 0.5 mL of water and then drying over potassium carbonate (~3 g) afforded ~380 g of an oligomeric mixture suitable for use in making diesel and jet fuels with flash points over 61 deg C.

[0034] **Figure 1** shows a beaker removed directly from SS-bomb showing a typical homogeneous oligomeric mixture from 1-butene that contains the Cp_2ZrCl_2/MAO catalyst system prepared in cyclohexane. The sample contains no aromatic hydrocarbons. It is a clear solution.

[0035] Where a range of values is provided, it is understood that each intervening value, to the tenth of the unit of the lower limit unless the context clearly dictates otherwise, between the upper and lower limits of that range is also specifically disclosed. Each smaller range between any stated value or intervening value in a stated range and any other stated or intervening value in that stated range is encompassed within the invention. The upper and lower limits of these smaller ranges may independently be included or excluded in the range, and each range where either, neither or both

limits are included in the smaller ranges is also encompassed within the invention, subject to any specifically excluded limit in the stated range. Where the stated range includes one or both of the limits, ranges excluding either or both of those included limits are also included in the invention.

CLAIMS

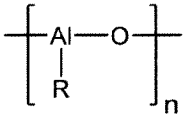
What is Claimed is:

1. A process for preparing an Activated Homogeneous Metallocene Catalyst (AHMC) suited for use in the dimerization and oligomerization of α-olefins, comprising:

contacting a metallocene precatalyst of the formula (I):

$L^1L^2MX^1X^2$ (I)

with an alkylaluminoxane of formula (II):



(II); and

at least one of the precatalyst, the alkylaluminoxane, and their combination being combined with at least one saturated hydrocarbon solvent,

wherein:

M is Ti, Zr, or Hf;

X^1 and X^2 are independently at least one of halogen, hydrogen, alkyl, and a mixture thereof;

L^1 and L^2 independently comprise at least one π-ligand selected from the group consisting of cyclopentadienyl, indenyl, fluorenyl, and derivatives thereof,

n is an integer of at least 3;

R comprises a C_1-C_{30} alkyl; and

the at least one saturated hydrocarbon solvent optionally being selected from the group consisting of cycloalkanes, normal alkanes, iso-alkanes, and combinations thereof.

2. The process according to claim 1, further characterized by combining the precatalyst with the at least one saturated hydrocarbon solvent prior to contacting it with the alkylaluminoxane.

3. The process according to claim 1, further characterized by said π-ligand being a derivative of cyclopentadienyl, indenyl, or fluorenyl, and wherein substituents are attached to said ligands through ansa-linkages that bind the respective rings, with at least one carbon or silicon atom, each bearing hydrogen or alkyl radicals.

4. The process according to claim 1, further characterized by R being a linear or branched alkyl, or a combination thereof.

5. The process according to claim 1, further characterized by the contacting including an Al/M molar ratio of from 1 to 100.

6. The process according to claim 5, further characterized by said Al/M molar ratio being from 10 to 20.

7. A process for oligomerizing an olefin characterized by:

contacting an olefin feedstock with the Activated Homogeneous Metallocene Catalyst prepared according to the process of any one of claims 1-6 to form an olefinic-oligomer product, the olefin feedstock comprising at least one olefin,

the olefin optionally comprising an α-olefin.

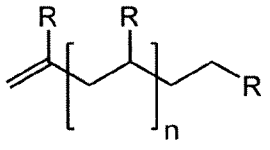
8. The process of claim 7, further characterized by the olefin feedstock comprising less than 20 wt. %, or less than 10 wt. % or less than 5 wt. % or less than 1 wt. % of all constituents other than the at least one olefin.

9. The process of claim 7, further characterized by the contacting including contacting the olefin feedstock for at least one hour.

10. The process of claim 7, further characterized by the olefin comprising an α-olefin and the contacting forming an α-olefinic-oligomer in at least a 98% conversion.

11. The process of claim 7, further characterized by the olefin comprising an α-olefin

and the contacting forming an α-olefinic-oligomer of according to formula (III):



(III)

12. The process according to claim 7, further characterized by said AHMC being introduced in one portion, in discrete aliquots, or through a continual addition process directly to the olefin feedstock.

13. The process according to claim 7, further characterized by a ratio of metallocene precatalyst (g of M) to α-olefinic-oligomer product (Kg) being from 5 to 40.

14. The process according to claim 7, further characterized by transferring said olefinic-oligomer product to a quench vessel, treated with water, and then filtered to remove solids.

15. The process according to claim 7, further characterized by passing said olefinic-oligomer product through metal oxide filter material to remove M and Al species from said product solution.

16. The process according to claim 15, further characterized by the metal oxide comprising alumina.

17. The process according to claim 7, further characterized by treating said olefinic-oligomer product with a stabilizing agent selected from a hydroquinone or phenol family of stabilizers, optionally with butylated hydroxyl toluene (BHT), at a concentration of 10-300 ppm.

18. The process according to claim 7, further characterized by removing, by flash distillation, a lower boiling fraction of the olefinic-oligomer products.

19. The process according to claim 7, further characterized by removing a lower boiling fraction of α-olefinic-oligomer products between 50 °C and 150 °C, or between 70 °C and 130 °C.

20. The process according to claim 18 or 19, further characterized by said lower boiling fraction being from 25 to 45 wt. % of the total α-olefinic-oligomer product, or 30-40 wt. % of the total α-olefinic-oligomer product.

21. The process according to claim 7, further characterized by subjecting a light boiling fraction of said α -olefinic-oligomer product to selective-dimerization to create a mixture of mono-olefinic hydrocarbon products having a boiling point greater than 200 °C but less than 300 °C at atmospheric pressure.

22. The process according to claim 7, further characterized by subjecting a high boiling fraction of said α-olefinic-oligomer product to catalytic hydrogenation over at least one heterogeneous metal catalyst comprising at least one of palladium, platinum, and nickel.

23. The process according to claim 22, further characterized by said subjecting being performed at a hydrogen pressure in the range of 300-20800 kpa, or between 440 and790 kpa.

24. The process according to claim 22, further characterized by said subjecting being performed at a temperature in the range of 20 to 100 °C, or at 20-50°C, or at 35-45°C, or at 60-80°C.

25. The process according to claim 22, further characterized by said product being subjected to hydrogenation over a heterogeneous metal catalyst which comprises at least one of palladium, platinum, and nickel and hydrogen pressure in the range of 300-20800 kpa, or a pressure between 7000 and 14000 kpa.

26. The process according to claim 25, further characterized by said products being combined and distilled at atmospheric pressure.

27. The process according to claim 25, further characterized by said products obtained being combined and distilled at reduced pressure.

28. The process according to claim 26, further characterized by collecting a hydrocarbon product, a completely 100% synthetic iso-paraffinic kerosene (SiPK) product, starting at 150 °C and finishing at 280 °C, suitable as a jet/diesel fuel.

29. The process according to claim 26, further characterized by the product collected providing a fuel mixture that meets Jet-A/JP-8 and diesel #1 flashpoint requirements (≥38 °C).

30. The process according to claim 28 or 29, further characterized by said SiPK jet/diesel fuel produced having a derived Cetane index of 40-55, or a Cetane index of 50.

31. The process according to claim 28 or 29, further characterized by said SiPK jet/diesel fuel produced having a maximum cold flow viscosity of 8 cSt as measured at -20 °C (ASTM 445LT).

32. The process according to claim 26, further characterized by collecting a product starting at 170 °C and finishing at 280 °C which yields a SiPK jet/diesel that meets military JP-5 and diesel #2 flashpoint requirements with a flashpoint >61 °C.

33. The process according to claim 32, further characterized by said SiPK jet/diesel fuel produced having a derived Cetane index of 40-55 or 50 Cetane index.

34. The process according to claim 32 or 33, further characterized by said SiPK jet/diesel fuel produced having a maximum cold flow viscosity of 8.5 cSt as measured at -20 °C (ASTM 445LT).

35. The process according to claim 7, further characterized by said olefin feedstock comprising an α-olefin selected from the group consisting of 1-propene, 1-butene, 1-pentene, 1-hexene, and combinations thereof.

36. The process according to claim 35, further characterized by said α-olefin being neat 1-butene or a mixture of 1-butene and 1-propene, optionally at a ratio of 1-butene to 1-propene of 3:1, mol:mol.

37. The process according to claim 7, further characterized by the contacting being performed in the absence of aromatic solvents.

38. A fuel produced by the method of any one of claims 7-19, 21-29, 32-33, and 35-37.

39. The process according to claim 1, further characterized by said aliphatic hydrocarbon solvent is a cycloaliphatic hydrocarbon.

40. The process according to claim 1, further characterized by said π-ligands are cyclopentadienyl, R is methyl, and said aliphatic hydrocarbon solvent is at least one of cyclohexane and methylcyclohexane.

41. The process according to claim 1, further characterized by said π-ligands are cyclopentadienyl rings having at least one alkyl group attached, R is methyl, and the aliphatic hydrocarbon solvent is selected from the group consisting of cyclic, acyclic linear and branched hydrocarbon solvents that include jet and diesel fuels.

42. The process according to claim 1, further characterized by said π-ligands are cyclopentadienyl, R is at least three carbons, and the aliphatic hydrocarbon solvent is selected from acyclic linear and/or branched hydrocarbon solutions and fuels.

43. The process according to claim 1, further characterized by said alkylaluminoxane being prepared by direct reaction of one mole of trialkylalane with one mol-equivalent of water in iso-paraffinic kerosene.

44. The process according to claim 43, further characterized by said trialkylalane

being selected from the group consisting of trimethylalane, triethylalane, tripropylalane, tri-isopropylalane, tributylalane, tri-isobutylalane, tri-isobutylalane and combinations thereof.

45. The process according to claim 1, further characterized by said π-ligands are cyclopentadienyl rings having at least two alkyl groups attached, R is methyl, and the aliphatic hydrocarbon solvent is selected from the group consisting of cyclic, acyclic linear, branched hydrocarbon solvent, and jet and diesel fuels prepared.

46. The process according to claim 1, further characterized by said solvent being a fuel having a flashpoint of at least 61, or 61- 100.

47. The process according to claim 1, further characterized by an isomerization catalyst at from 0.1 ppm to 0.1 weight %, relative to the total olefin component.

48. An Activated Homogeneous Metallocene Catalyst produced by the process of any one of claims 1-6 and 39-47.

49. A process for forming a fuel, comprising:

providing an olefin feedstock comprising at least of an one alpha-olefin and an internal olefin;

combining the olefin feedstock with a catalyst comprising at least one homogenous Ziegler-Natta catalyst and at least one homogenous activating co-catalyst, and at least one hydrocarbon solvent to form the fuel by at least one of oligomerization and dimerization.

50. The process according to claim 49, further characterized by the combining being performed in the absence of any aromatic solvents.

51. The process according to claim 49, further characterized by at least one olefin-isomerization catalyst.

52. The process according to claim 49, further characterized by said Ziegler-Natta

catalyst including a group four (4) metallocene catalyst.

53. The process according to claim 49, wherein said co-catalyst is an alkylaluminoxane (AAO).

54. The process according to claim 49, further characterized by said catalyst being prepared by contacting a metallocene precatalyst with an aliphatic hydrocarbon solution having an alkylaluminoxane (AAO).

55. The process according to claim 49, further characterized by said aliphatic hydrocarbon solvent being derived from a fuel.

56. The process according to claim 49, further characterized by said catalyst being prepared by dissolving a trialkylalane in an aliphatic-hydrocarbon solvent and/or fuel and treating it with one mol-equivalent of water, which is then contacted with a metallocene precatalyst and filtered.

57. The process according to claim 55, further characterized by said trialkylalanes being selected from the group consisting of trimethylalane, triethylalane, tributylalane, and tri(iso-butyl)alane (TIBA).

58. The process according to claim 55, further characterized by said aliphatic hydrocarbon solvents and/or fuels being selected from the group consisting of straight chain alkanes including hexanes, heptanes, octanes, nonanes, decanes, and alkanes having greater than 10 carbons.

59. The process according to claim 55, further characterized by said aliphatic hydrocarbon solvents and/or fuels being selected from the group consisting of branched-aliphatic-hydrocarbons of 6 to 16 carbons.

60. The process according to claim 59, further characterized by said branched-

aliphatic-hydrocarbon being selected from the group consisting of 3-methylheptane, 2-methyloctane, and combinations thereof.

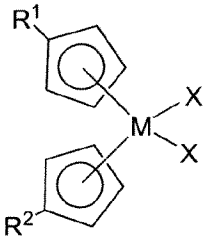
61. The process according to claim 55, further characterized by said aliphatic hydrocarbon solvents and/or fuels being selected from the group consisting of cyclic aliphatic-hydrocarbons including cyclohexanes, methylcyclohexanes, dimethylcyclohexanes, tetralins, pinanes, and other mono- and bicyclic aliphatic-hydrocarbons.

62. The process according to claim 49, further characterized by said fuel having a flashpoint of 61 to 100 °C.

63. A fuel prepared by the process of any one of claims 49-62.

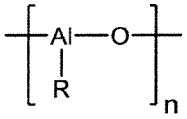
64. A process for oligomerizing an olefin characterized by:

a) contacting a metallocene precatalyst of the formula (I):



(I)

with an alkylaluminoxane of formula (II):



(II);

the precatalyst contacted with the alkylaluminoxane being combined with at least one saturated hydrocarbon solvent to provide an Activated Homogeneous Metallocene Catalyst,

wherein:

M is Ti, Zr, or Hf;

X^1 and X^2 are independently at least one of halogen, hydrogen, alkyl, and a mixture thereof;

R comprises a C_1-C_{30} alkyl;

R^1 and R^2 are independently comprise a C_1-C_{30} alkyl or H,

n is an integer of at least 3;

; and

the at least one saturated hydrocarbon solvent is selected from the group consisting of cycloalkanes, normal alkanes, iso-alkanes, and combinations thereof; and

b) contacting an olefin feedstock with the Activated Homogeneous Metallocene Catalyst form an olefinic-oligomer product, the olefin feedstock comprising at least one olefin.

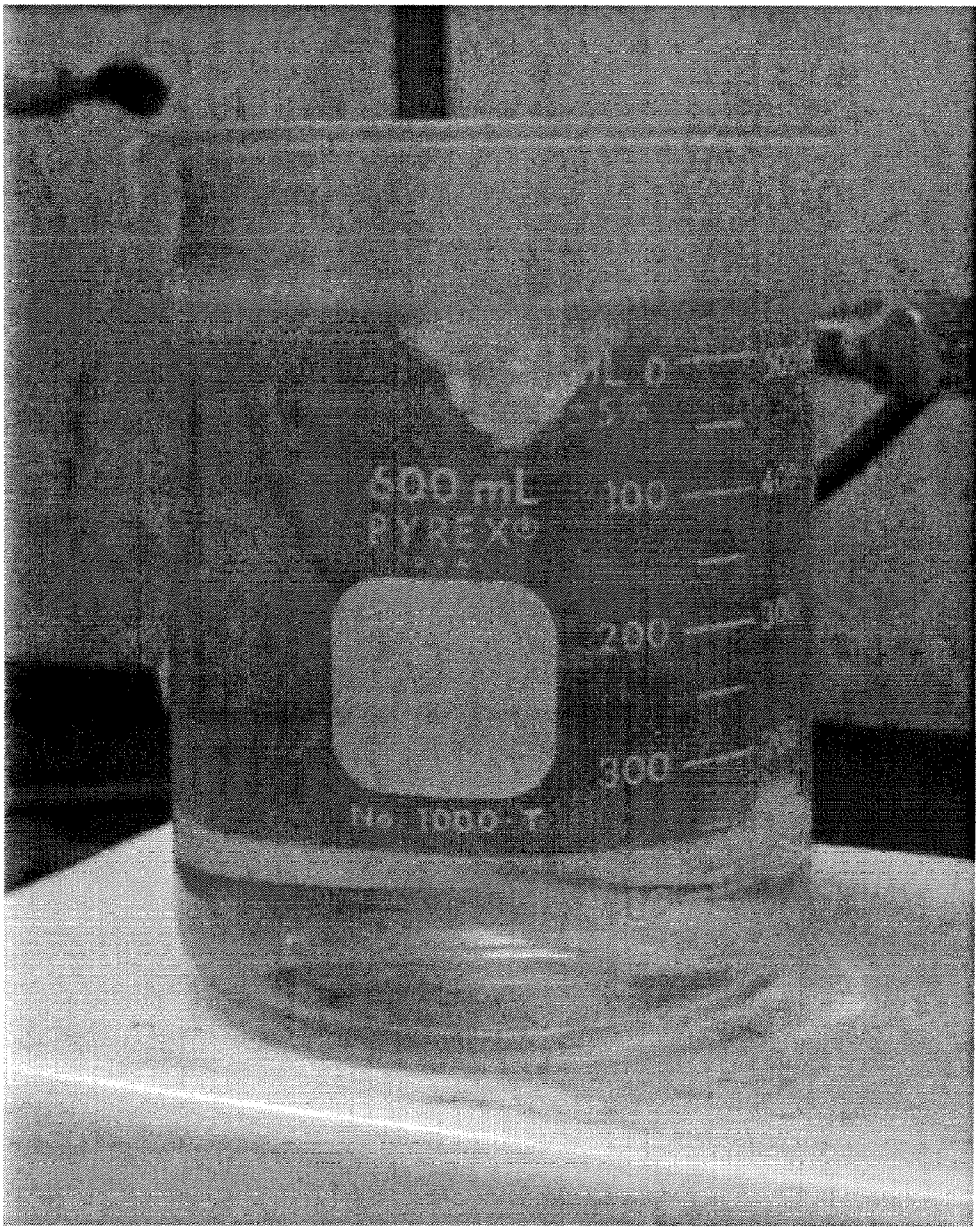


FIGURE 1

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US 12/35167

A. CLASSIFICATION OF SUBJECT MATTER
IPC(8) - B01J 31/00; B01J 37/00 (2012.01)
USPC - 502/104
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC(8) -B01J 31/00; B01J 37/00 (2012.01)
USPC -502/104

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
USPC - 502/118, 152; 526/160, 165, 943 (see search terms below)

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
PubWEST, Google Patents, Google Scholar, WIPO (Metallocene, precatalyst, catalyst, process, formula, aluminoxane, olefin, homogeneous, aluminoxane, ligand, kerosene, Ziegler, quench, flash, point, oligomer, activated, alkylaluminoxane, fuel, solvent, diesel, metal, oxide, alumina, dimerization, filter, medium, internal, natta, co-catalyst

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X --- Y	US 2007/0293640 A1 (JIANG, et al.) 20 December 2007 (20.12.2007) entire document, especially para[0007], [0020], [0038]-[0041], [0058], [0096], [0112]-[0113], [0122], [0128], [0135], [0141]-[0144], [0162], [0219], [0246]-[0250], [0255], [0260]-[0263], [0266]-[0267], [0962], [0977], [1038], [1070], [1187], [1189].	1-14,19,20/19,35-37, 39-40,42,47,48/(1-6,39-42, 47), 64 --- 15-18, 20/18, 21-34, 41, 43-46, 48/(41,43-46), 49-63
Y	US 2007/0185362 A1 (LATTNER, et al.) 09 August 2007 (09.08.2007) entire document, especially para[003], [0344], [0354].	15-16, 18, 20/18
Y	US 5,158,992 A (CASSELLI, et al.) 27 October 1992 (27.10.1992) entire document, especially col 1, ln 54-56; col 5, ln 52-55.	17 21
Y	US 2007/0293712 A1 (TITTA, et al.) 20 December 2007 (20.12.2007) entire document, especially para[0033], [0044].	22-34
Y	US 2011/0114538 A1 (COSYNS, et al.) 19 May 2011 (19.05.2011) entire document, especially para[0050]-[0051], [0054], [0058], [0126].	31-34, 41, 45-46, 48/(41,45-46), 49-62, 63/(49-62)
Y	US 2009/0305926 A1 (WU, et al.) 10 December 2009 (10.12.2009) entire document, especial para[0001], [0004], [0007]-[0008], [0018]-[0019], [0029], [0035], [0050]-[0051], [0072], [0090], [0126].	43-44, 48/(43-44), 56-57, 63/(56-57)
Y	US 4,772,736 A (EDWARDS, et al.) 20 September 1988 (20.09.1988) entire document, especially col 1, ln 27-col 2, ln 2.	

☐ Further documents are listed in the continuation of Box C. ☐

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" earlier application or patent but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
05 August 2012 (05.08.2012)	14 AUG 2012
Name and mailing address of the ISA/US Mail Stop PCT, Attn: ISA/US, Commissioner for Patents P.O. Box 1450, Alexandria, Virginia 22313-1450 Facsimile No. 571-273-3201	Authorized officer: Lee W. Young PCT Helpdesk: 571-272-4300 PCT OSP: 571-272-7774

Form PCT/ISA/210 (second sheet) (July 2009)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US 12/35167

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. ☒ Claims Nos.: 38
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying additional fees, this Authority did not invite payment of additional fees.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.

☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.

☐ No protest accompanied the payment of additional search fees.

Form PCT/ISA/210 (continuation of first sheet (2)) (July 2009)

(19)中华人民共和国国家知识产权局

(12)发明专利申请

(10)申请公布号 CN 104926600 A
(43)申请公布日 2015.09.23

(21)申请号 201510239573.4

(22)申请日 2015.05.12

(71)申请人 佛山市德品高新材料有限公司
地址 528306 广东省佛山市顺德区容桂镇容里建丰路7号

(72)发明人 陈健 胡学一 方云

(51)Int.Cl.
C07C 29/03(2006.01)
C07C 31/20(2006.01)

权利要求书1页 说明书8页 附图1页

(54)发明名称

一种一锅法合成1,2-戊二醇的方法

(57)摘要

本发明涉及一种低温条件下由正戊烯为原料一锅法高产率合成1,2-戊二醇的方法。本发明的反应温度低,降低了1-戊烯原料高温挥发而导致泄露风险,同时降低了对设备耐压需求;采用多步一锅法工艺缩短了工艺流程;反应中不采用有腐蚀、有污染和有害化合物为原料,消除传统工艺采用原料多倍量的有机酸载氧剂,该工艺的环境友好性佳;本发明的原料来源广泛,生产成本低,经济性好。

CN 104926600 A

1. 一种一锅法合成 1,2- 戊二醇的方法，其特征在于以 1- 戊烯、双氧水、催化剂在溶剂中加热到一定的温度反应下反应一定时间。

2. 权利 1 所述的双氧水，其特征在于过氧化氢浓度为 10%-80% (W/W)。

3. 权利 1 或权利 2 所述的 1- 戊烯与过氧化氢的物质量比，其特征在于比值为 1:1-2。

4. 权利 1 所述的催化剂，其特征在于催化剂为四氯化钛，或具有 $Ti(OR)_4$ 通式的钛酸酯，R 为 C_1-C_{10} 的烷基。

5. 权利 1 或权利 4 所述的催化剂，其特征在于加入量为原料 1- 戊烯质量的 0.1%-10% (W/W)。

6. 权利 1 所述的反应温度，其特征为范围为 10 ℃-100℃。

7. 权利 1 或权利 6 所述的反应温度，其特征为恒温反应。

8. 权利 1 或权利 6 所述的反应温度，其特征在于程序升温反应。

9. 权利 1 所述的反应时间为 1h-24h。

10. 权利 1 所述的溶剂，其特征在于溶剂为甲醇、乙醇、异丙醇、正丁醇、丙酮、甲乙酮、二丁酮中的一种或几种的混合物。

一种一锅法合成 1,2-戊二醇的方法

技术领域

[0001] 本发明涉及一种一锅法合成 1,2- 戊二醇的方法，属于有机化合物合成领域。

背景技术

[0002] 1,2- 戊二醇是合成高效、低毒和活性谱广的丙环唑类杀菌剂（比利时杨森医药集团[1]开发，已在多国获登记）的重要中间体，用于构建丙环唑分子的缩酮结构[2]。1,2- 戊二醇还具有保湿功能，与其它保湿剂复配使用产生较强的协同效应[3]，可用作化妆品或个人护理用品[3-4]的保湿功效成分。1,2- 戊二醇还具有抑菌性[5]，在不添加其它防腐剂的配方中亦可有效抑制产品的变质[6]。

[0003] 1,2- 戊二醇的合成方法主要以下两种：(1) 正戊烯环氧化反应和后继的水解反应；(2)糠醛或糠醇的加氢还原。第一种方法主要存在生产安全和污染大等不足；第二种方法主要存在原料转化率不高、副反应多和产品提纯困难等不足；具体情况见如下分析。

[0004] 德固赛公司[7]公开了以正戊烯为原料、苯为溶剂、丙酸为载氧剂和氢氧化钠为开环试剂的连续相中合成 1,2- 戊二醇的方法。Ciba-Geigy 公司公开了一种采用正戊烯和甲酸为原料，过氧化氢为氧化剂，氢氧化钠为开环试剂的合成 1,2- 戊二醇连续生产工艺[8]。浙江联盛集团[9]和中国石化[10]也公开了以正戊烯、甲酸和过氧化氢为原料的类似工艺。该工艺原料沸点低（正戊烯的沸点 30.1℃），易因低沸点有机化合物挥发导致的劳动者安全和生产安全等隐患；以甲酸为原料对设备的腐蚀性强，少量泄露的甲酸对操作者的皮肤有强腐蚀作用，同时以正戊烯物质的量 2-10 倍甲酸导致大量液体有机废弃物待处理；该反应的副产物多，产品的分离纯化相对困难。大连化物所[11]公开了以磷钨酸催化正戊烯与过氧化氢反应直接制备 1,2- 脂肪二醇的方法，解决了因用过量甲酸引起的一系列问题；但仍旧不能解决在反应条件下低沸点原料挥发和对设备耐压要求高等不足。中科院煤炭所朱玉雷[12]报道了合成 1,2- 戊二醇的绿色化学新途径，以二氧化锰负载贵金属钌催化糠醇加氢合成 1,2- 戊二醇。该方法中应用了贵金属钌，价格昂贵；同时催化剂的载体二氧化锰并不是特别适用于工业应用的载体材料。天津医科大学侯薇[13]报道了正戊烯为原料，硼酸钠为氧化剂，醋酸酐为反应溶剂和反应物合成了乙酸单戊二醇酯，该酯类化合物在碱性条件下进一步水解得 1,2- 戊二醇。该方法避免了甲酸等腐蚀性强的化合物。但是过量醋酸酐和氧化剂导致反应产品后处理困难；正戊烯与醋酸酐化学计量比反应生成乙酸单戊二醇酯，消耗了大量的乙酸酐，增加了原料成本；氧化反应形成单酯与水解反应的分离，延长了反应路线，增加了操作成本。Severeyns 等[14]以 NMO 和四氧化锇催化剂的作用下催化正戊烯的双羟基化反应合成 1,2- 戊二醇应的二醇。该方法使用的四氧化锇催化剂毒性高、价格昂贵、受热易产生有毒气体，因此该路线的工业应用价值极微。

[0005] 早在 1931 年，Adkins[15]报道了亚铬酸铜催化糠醇加氢合成了 1,2- 戊二醇和 1,5- 戊二醇的混合物，0.75mol 糠醇中加入 3g 催化剂，在 175℃下反应 11 小时，1,2- 戊二醇和 1,5 戊二醇的总产率最高达到 70%。但是 Manly 在 1958 年[16]采用 Adkins 相似的条件反应，1,2- 戊二醇的收率却仅仅为 4.4%。日本筑波大学的 Tomishige[17]在 Chem Comm 上发表了

金属负载的钌和铼催化糠醇的加氢还原反应，结果产物中未检测到1,2-戊二醇，而是选择性的生成了1,5-戊二醇。浙江大学许彦娟[18]研究了Cu-Zn、Cu-Mn、Cu-Cr、Raney Cu、Raney Ni及贵金属负载型催化剂Pt/C、Pd/C、Ru/ AI2O3等催化糠醇选择性加氢还原为1,2-戊二醇，其中Cu-Zn的催化性能最优。在反应温为150~160 ℃, 反应压力7~8MPa, 反应时间10 h下，糠醇转化率达到69.2%，1,2-戊二醇的选择性达到46.2 %, 其收率达到32.0%。原料转化率低、副反应多和1,2-戊二醇的收率低仍旧是该路线的致命缺陷。赢创工业集团[19]公开了糠醇加氢还原合成1,2-戊二醇的方法。珠海凯美科技有限公司[20]公开了非贵金属催化剂选择性加氢合成1,2-戊二醇。该方法利用农副产品深加工产物糠醛为原料，大大解决了受正戊烯原料限制等问题；该方法采用氧化铜等非贵金属催化剂催化加氢，解决了贵金属催化剂价格昂贵和回收困难等问题；但是该方法仍旧存在原料转化不完全，副反应多和产率低等问题，由此带来了分离困难、产品纯度不高和反应路线非环境友好等新问题；氧化铜催化加氢的性能差，导致加氢反应的压力过高，由此带来生产过程中的安全隐患。日本仙台紧凑型化工研究中心的Maya Chatterjee[21]报道了在超临界二氧化碳中催化四氢糠醇合成1,2-戊二醇，其中Pd/MCM-41为催化剂的1,2-戊二醇得率最高，达到77.4%，还含有12.6%的1,5-戊二醇和10%的1-戊醇和2-戊醇的混合物。该方法虽然提高了1,2-戊二醇的收率，但是仍旧采用负载型贵金属为催化剂，操作成本相对较高；采用超临界二氧化碳为溶剂，反应体系的压力也较大，该工艺也不适合于工业规模生产1,2-戊二醇。

[0006] 除了上述的两种方法外，还有一些零星报道合成1,2-戊二醇的方法。德固赛公司[22]公开了正丁醛与氢氰酸亲核反应生成邻羟基正戊腈，水解邻羟基正戊腈得到邻羟基正戊醇，再在贵金属钌和铼等非均相催化剂催化下合成1,2-戊二醇。该方法中原料氢氰酸为剧毒化学品，对操作者有潜在的致命性；反应分三步进行，路线长；第一步亲核反应因羟醛缩合等副反应使得反应控制困难，副产物多，产品提纯困难；使用贵金属钌和铼为催化剂，增加了生产成本。江苏扬农化工集团[23]公开了2-氯代戊醇在碱性溶剂中发生环氧化反应。该工艺虽可能高产率获得1,2-戊二醇，但2-氯代戊醇原料不易得，同时反应过程中常压蒸馏环氧戊烷尚存在操作不安全的致命隐患。江苏化工农药集团丁小兵在《安徽化工》报道了[24]用硼氢化钠还原邻羟基正戊酸合成1,2-戊二醇。硼氢化钠的价格昂贵和羟基正戊酸的不易得导致该工艺实现工业化生产困难重重。

[0007] 综上所述，已见文献报道的以正戊烯环氧化反应、环氧化合物开环反应合成1,2戊二醇的路线存在原料沸点低和挥发性强、该工艺产生的液体废弃物多、产品的分离和后处理困难等不足；已见文献报道的以糠醇或糠醛加氢还原反应合成1,2-戊二醇的路线虽然解决了原料来源问题，但仍旧存在贵金属催化剂价昂、原料转化率不高、产物选择性不佳和产品纯度低等不足；报道的其它方法或存在原料价昂、或原料来源不足、或反应过程中使用到剧毒化学品、或路线长、或反应困难、或产物提纯困难等不足。

[0008] [背景技术的参考文献]

1. Van Reet, G.; Heeres, J.; Wals, L. Antimicrobial and plant-growth-regulating triazole derivatives. US4160838, 19780103., 1979.

2. (a) Palecek, J.; Kubelka, V.; Dedek, V.; Mostecky, J. 1-Substituted imidazoles and triazoles. CS215362, 19800522., 1984; (b) Lourens, G. J.; Booysen, M.; Joubert, E. preparation of propiconazole precursor 2-bromomethy

1-2-(2,4-dichlorophenyl)-4-propyldioxolane via bromination of 2-(2,4-dichlorophenyl)-2-methyl-4-propyldioxolane. ZA9604976, 19960612., 1997; (c) Darpan, P.; Prasanna, K. A process for the preparation of unsymmetrical triazole fungicides. IN2006MU00697, 20060504., 2008.

3. Schmaus, G.; Pillai, R. Use of synergistically active 1, 2-alkanediol mixtures as skin moisture-regulating compositions. WO2006069953, 20051221., 2006.

4. (a) Schrader, K. Alkanediols as skin moisturizers. DE4320744, 19930623., 1995; (b) Serizawa, S. Cosmetic moisturizers containing bisabolol and polyvalent alcohols. JP11060463, 19970819., 1999; (c) Kuroda, A.; Iida, M.; Nakamura, T. Skin-moisturizing cosmetic compositions containing 1,2-pentanediol and isoprene glycol. JP2002047168, 20000801., 2002; (d) Omori, T.; Miyahara, R.; Kanokogi, H.; Nanba, T. Preparation of alkyl polyglycol neopentanoates and their use as moisturizers, cosmetics, and topical use. JP2002302469, 20010330., 2002; (e) Iida, T.; Yamaguchi, F.; Tadenuma, H. Liquid dishwashing detergent compositions and skin-moisturizing method using them. JP2006193732, 20051214., 2006.

5. Roeding, J.; Lange, S.; Schmaus, G.; Joppe, H.; Pillai, R. Synergistic mixtures of at least one 1,2 alkanediol such as 1,2-hexanediol and 1,2-octanediol with a further compound having antimicrobial properties. US20070265352, 20051020., 2007.

6. (a) Sumino, H.; Asaka, Y. Zinc gluconate and 2-phenoxyethanol or 1,2-pentanediol as antimicrobials for cosmetics. JP2004067626, 20020808., 2004; (b) Merianos, J. J.; Garelick, P.; Lindstrom, S. M. Antimicrobial compositions containing diols. US20050228032, 20040408., 2005; (c) Faergemann, J.; Hedner, T. Antimicrobial compositions comprising at least three different diols. WO2006057616, 20051129., 2006; (d) Tominaga, H.; Yamamoto, Y.; Ohashi, Y. Antimicrobial cosmetics or skin preparations containing alkyl glucosides and specific antibacterial agents. JP2010265189, 20090513., 2010.

7. Siegmeier, R.; Prescher, G.; Maurer, H.; Hering, G. Continuous production of 1,2-pentanediol. DE3412937, 19841124., 1986.

8. Issler, H.; Maul, R. Continuous process for producing 1,2-alkanediols. US4479021, 19830830., 1984.

9. 俞小鸥 一种1,2-戊二醇的制备方法. CN1552684, 2005.

10. 傅建松; 王继媛; 孙超; 张冬梅; 吴忠平 一种制备1,2-戊二醇的方法. CN103864575A, 2012.

11. 高爽; 吕迎; 张恒耘; 奚祖威 一种1,2-脂肪二醇的制备方法. CN1923777, 2005.

12. Zhang, B.; Zhu, Y.; Ding, G.; Zheng, H.; Li, Y., Selective conversion of

furfuryl alcohol to 1,2-pentanediol over a Ru/MnOx catalyst in aqueous phase. *Green Chemistry* 2012,*14* (12), 3402-3409.

13. 侯薇；刘继华 用乙酐合成法制备邻二醇．1996.

14. Severeyns, A.; De Vos, D. E.; Fiermans, L.; Verpoort, F.; Grobet, P. J.; Jacobs, P. A., A heterogeneous cis-dihydroxylation catalyst with stable, site-isolated osmium-diolate reaction centers. *Angewandte Chemie, International Edition* 2001,*40* (3), 586-589.

15. Adkins, H.; Connor, R., Catalytic hydrogenation of organic compounds over copper chromite. *Journal of the American Chemical Society* 1931,*53*, 1091-5.

16. Manly, D. G.; Dunlop, A. P., Catalytic hydrogenation. I. Kinetics and catalyst composition in the preparation of 2-methylfuran. *Journal of Organic Chemistry* 1958,*23*, 1093-5.

17. Koso, S.; Furikado, I.; Shimao, A.; Miyazawa, T.; Kunimori, K.; Tomishige, K., Chemoselective hydrogenolysis of tetrahydrofurfuryl alcohol to 1,5-pentanediol. *Chemical Communications (Cambridge, United Kingdom)* 2009, (15), 2035-2037.

18. 许彦娟 糠醇催化加氢制备 1,2-戊二醇的研究．2014.

19. M. 奥迈斯；M. 诺伊曼；V. 布雷梅；C. 泰斯；D. 沃尔夫；P. 克劳斯；M. 卢卡斯；R. 埃克特 糠醇氢解成 1,2-戊二醇．CN104016831A.

20. 崔健；崔晓桐，糠醛一步加氢生产 1,2-戊二醇的方法．

21. Chatterjee, M.; Kawanami, H.; Ishizaka, T.; Sato, M.; Suzuki, T.; Suzuki, A., An attempt to achieve the direct hydrogenolysis of tetrahydrofurfuryl alcohol in supercritical carbon dioxide. *Catalysis Science & Technology* 2011,*1* (8), 1466-1471.

22. Krimmer, H.-P.; Sans, J.; Theis, C.; Thalhammer, F. Process for preparing 1,2-diols from carbonyl compounds via the corresponding cyanohydrins. US20080064905, 20070822., 2008.

23. 丁克鸿；程晓曦；郑亚清；许金来；吴惠；王根林；杨进；陆春明；金野 一种制备 1,2-戊二醇的新方法．CN101885668A, 2010.

24. 丁小兵，1,2-戊二醇的合成．*安徽化工* 2002,(05), 22-23+21.

发明内容

[0009] ［要解决的问题］

本发明的目的在于提供一种在低温条件下由正戊烯为原料合成 1,2-戊二醇的方法。

[0010] 本发明的另一个目的在于提供一种对设备无腐蚀的原料合成 1,2-戊二醇的方法。

[0011] 本发明的另一个目的在于提供一种对环境更加友好的由 1-戊烯为原料合成 1,2-戊二醇的方法。

[0012] ［技术方案］

本发明所述的方法，包括以下步骤：

1 在反应釜中分别加入一定量溶剂和催化剂，并混合均匀；

2 在反应釜中再加入一定量正戊烯，并混合均匀；

3 保持反应釜内物料温度为0-40 ℃，并在慢速搅拌条件下缓慢滴加一定量双氧水，反应数小时；

4 在反应釜中加入一定量水或不加水；

5 在反应釜中加入一定量的二氧化锰；

6 将反应釜内温度上升至30-80 ℃，反应数小时；

7 反应结束后蒸发体系中的溶剂，过滤后得1,2-戊二醇粗产品；

8 上述粗产品经减压蒸馏后得含量大于99.5%的1,2-戊二醇。

[0013] ［有益效果］

与现有1,2-戊二醇的合成方法相比，本发明的有益效果如下：

1 本发明提供的反应温度为低热，降低了1-戊烯原料高温挥发而导致泄露风险和降低对设备耐压需求。

[0014] 2 本发明提供的多步一锅法反应工艺路线短，加工简便和产品产率高的工艺路线合成1,2-戊二醇。

[0015] 3 本发明的工艺过程无腐蚀、无污染、不产生任何有害化合物，因此本发明的合成工艺有较好的环境友好性佳。

[0016] 4 本发明方法的原料来源广泛且低廉，工艺路线短，生产成本低，经济性好。

附图说明

[0017] 图1 为1,2-戊二醇的溴化钾盐片涂膜法的傅立叶变换红外光谱图。图中3361cm^{-1}处为缔合羟基的伸缩振动吸收；2958 cm^{-1}，2931 cm^{-1}和2872 cm^{-1}处为甲基和亚甲基的碳氢伸缩振动的特征吸收。

[0018] 图2 为实施例2的产品1,2-戊二醇的气相色谱图。气相色谱条件为：OV1701毛细管柱、30m、0.32um，氮气为载气、柱前压0.1 MPa，柱温180℃。面积归一化百分含量为99.89%，以丙二醇为内标物的内标标准曲线法测定的含量为99.85%。

具体实施方式

[0019] 下面结合实施例对本发明作出进一步说明，但本发明不限于以下实施例。

[0020] 实施例1

在具有加热、温控、磁力搅拌、恒压滴液和回流装置的常压反应器中分别加入20 mL甲醇和0.1 g 四氯化钛，慢速搅拌使之混合均匀；在慢速搅拌条件下用恒压滴液漏斗滴加入10 g正戊烯；在10 ℃条件下，在30 min左右经恒压分液漏斗缓慢滴加15 g 30%的过氧化氢水溶液，滴加完毕后在10 ℃下继续搅拌6 h；然后向反应器中再加入20 g水，再在50 ℃下反应8 h；在反应器中加入0.1 g二氧化锰，蒸馏回收溶剂甲醇并除去水分，过滤得10.3 g 纯度为97.3%（气相色谱，1,2-丙二醇内标物的内标标准曲线法，）的1,2戊二醇粗产品。

[0021] 实施例2

在具有加热、温控、磁力搅拌、恒压滴液和回流装置的常压反应器中分别加入20 mL甲

醇和 0.1 g 钛酸四甲酯，慢速搅拌使之混合均匀；在慢速搅拌条件下用恒压滴液漏斗滴加入 10 g 正戊烯；在 10 ℃条件下，在 30 min 左右经恒压分液漏斗缓慢滴加 14 g 浓度为 50% 的过氧化氢水溶液，滴加完毕后在 10 ℃下继续搅拌 6 h；然后向反应器中再加入 30 g 水，再在 50 ℃下反应 8 h；在反应器中加入 0.1 g 二氧化锰，蒸馏回收溶剂甲醇并除去水分，过滤得 13.0 g 纯度为 97.4%（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2 戊二醇粗产品；经减压蒸馏，收集 5 mmHg 下 80 ℃左右的馏分，得 12.1 g 纯度为 99.6 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2- 戊二醇产品。

[0022] 实施例 3

在具有加热、温控、磁力搅拌、恒压滴液和回流装置的常压反应器中分别加入 20 mL 异丙醇和 1.0 g 钛酸四丁酯，慢速搅拌使之混合均匀；在慢速搅拌条件下用恒压滴液漏斗滴加入 10 g 正戊烯；在 10 ℃条件下，在 30 min 左右经恒压分液漏斗缓慢滴加 40 g 浓度为 10% 的过氧化氢水溶液，滴加完毕后在 10 ℃下继续搅拌 8 h；将体系温度升高到 80 ℃，继续反应 8 h；在反应器中加入 0.1 g 二氧化锰，蒸馏回收溶剂异丙醇并除去水分，过滤得 12.4 g 纯度为 95.2%（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2 戊二醇粗产品。

[0023] 实施例 4

在具有加热、温控、磁力搅拌、恒压滴液和回流装置的常压反应器中分别加入 20 mL 异丙醇和 2.0 g 钛酸四异丙酯，慢速搅拌使之混合均匀；在慢速搅拌条件下用恒压滴液漏斗滴加入 10 g 正戊烯；在 10 ℃条件下，在 30 min 左右经恒压分液漏斗缓慢滴加 20 g 浓度为 20 % 的过氧化氢水溶液，滴加完毕后在 10 ℃下继续搅拌 10 h；然后向反应器中再加入 20 g 水，再在 60 ℃下反应 5 h；在反应器中加入 0.1 g 二氧化锰，蒸馏回收溶剂异丙醇并除去水分，过滤得 13.2 g 纯度为 96.5 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2 戊二醇粗产品。

[0024] 实施例 5

在具有加热、温控、磁力搅拌、恒压滴液和回流装置的常压反应器中分别加入 20 mL 乙醇和 1.5 g 钛酸四己酯，慢速搅拌使之混合均匀；在慢速搅拌条件下用恒压滴液漏斗滴加入 10 g 正戊烯；在 10 ℃条件下，在 30 min 左右经恒压分液漏斗缓慢滴加 8 g 浓度为 80 % 的过氧化氢水溶液，滴加完毕后在 40 ℃下继续搅拌 2 h；然后向反应器中再加入 20 g 水，再在 70 ℃下反应 4 h；在反应器中加入 0.1 g 二氧化锰，蒸馏回收溶剂乙醇并除去水分，过滤得 12.5 g 纯度为 95.3 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2 戊二醇粗产品；经减压蒸馏，收集 5 mmHg 下 80 ℃左右的馏分，得 11.2 g 纯度为 99.5 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2- 戊二醇产品。

[0025] 实施例 6

在具有加热、温控、磁力搅拌、恒压滴液和回流装置的常压反应器中分别加入 30 mL 丙酮和 1.0 g 钛酸四异己酯，慢速搅拌使之混合均匀；在慢速搅拌条件下用恒压滴液漏斗滴加入 10 g 正戊烯；在 10 ℃条件下，在 30 min 左右经恒压分液漏斗缓慢滴加 20 g 浓度为 30 % 的过氧化氢水溶液，滴加完毕后在 20 ℃下继续搅拌 4h；然后向反应器中再加入 10 g 水，再在 40 ℃下反应 20 h；在反应器中加入 0.1 g 二氧化锰，蒸馏回收溶剂丙酮并除去水分，过滤得 12.7 g 纯度为 95.9 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2 戊二醇粗产品。

[0026] 实施例 7

在具有加热、温控、磁力搅拌、恒压滴液和回流装置的常压反应器中分别加入 20 mL 甲乙酮和 0.3 g 四氯化钛，慢速搅拌使之混合均匀；在慢速搅拌条件下用恒压滴液漏斗滴加入 10 g 正戊烯；在 10 ℃条件下，在 30 min 左右经恒压分液漏斗缓慢滴加 12 g 浓度为 30 %的过氧化氢水溶液，滴加完毕后在 30 ℃下继续搅拌 2 h；然后向反应器中再加入 10 g 水，再在 50 ℃下反应 16 h；在反应器中加入 0.1 g 二氧化锰，蒸馏回收溶剂甲乙酮并除去水分，过滤得 11.2 g 纯度为 98.1 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2 戊二醇粗产品。

[0027] 实施例 8

在具有加热、温控、磁力搅拌、恒压滴液和回流装置的常压反应器中分别加入 20 mL 四氢呋喃和 0.2 g 钛酸四甲酯，慢速搅拌使之混合均匀；在慢速搅拌条件下用恒压滴液漏斗滴加入 10 g 正戊烯；在 10 ℃条件下，在 30 min 左右经恒压分液漏斗缓慢滴加 15 g 浓度为 30 %的过氧化氢水溶液，滴加完毕后在 40 ℃下继续搅拌 1 h；然后向反应器中再加入 20 g 水，再在 50 ℃下反应 4 h；在反应器中加入 0.1 g 二氧化锰，蒸馏回收溶剂四氢呋喃并除去水分，过滤得 14.5 g 纯度为 98.3 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2 戊二醇粗产品；经减压蒸馏，收集 5 mmHg 下 80 ℃左右的馏分，得 13.5 g 纯度为 99.9 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2- 戊二醇产品。

[0028] 实施例 9

在具有加热、温控、磁力搅拌、恒压滴液和回流装置的常压反应器中分别加入 20 mL 二乙酮和 0.3 g 钛酸四乙酯，慢速搅拌使之混合均匀；在慢速搅拌条件下用恒压滴液漏斗滴加入 10 g 正戊烯；在 10 ℃条件下，在 30 min 左右经恒压分液漏斗缓慢滴加 10 g 浓度为 50 %的过氧化氢水溶液，滴加完毕后在 20 ℃下继续搅拌 10 h；然后向反应器中再加入 20 g 水，再在 50 ℃下反应 4 h；在反应器中加入 0.1 g 二氧化锰，蒸馏回收溶剂四氢呋喃并除去水分，过滤得 13.7 g 纯度为 98.0 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2 戊二醇粗产品；经减压蒸馏，收集 5 mmHg 下 80 ℃左右的馏分，得 12.4 g 纯度为 99.7 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2- 戊二醇产品。

[0029] 实施例 10

在具有加热、温控、磁力搅拌、恒压滴液和回流装置的常压反应器中分别加入 200 mL 无水甲醇和 3.0 g 钛酸四甲酯，慢速搅拌使之混合均匀；在慢速搅拌条件下用恒压滴液漏斗滴加入 200 g 正戊烯；在 10 ℃条件下，在 30 min 左右经恒压分液漏斗缓慢滴加 300 g 浓度为 30 %的过氧化氢水溶液，滴加完毕后在 10 ℃下继续搅拌 10 h；然后向反应器中再加入 200 g 水，再在 50 ℃下反应 14 h；在反应器中加入 1 g 二氧化锰，蒸馏回收溶剂甲醇并除去水分，过滤得 290.2 g 纯度为 98.1 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2 戊二醇粗产品；经减压蒸馏，收集 5 mmlg 下 80 ℃左右的馏分，得 281.0 g 纯度为 99.7 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2- 戊二醇产品。

[0030] 实施例 11

在具有加热、温控、磁力搅拌、恒压滴液和回流装置的常压反应器中分别加入 200 mL 四氢呋喃和 3.0 g 钛酸四甲酯，慢速搅拌使之混合均匀；在慢速搅拌条件下用恒压滴液漏斗滴加入 200 g 正戊烯；在 10 ℃条件下，在 30 min 左右经恒压分液漏斗缓慢滴加 150 g 浓

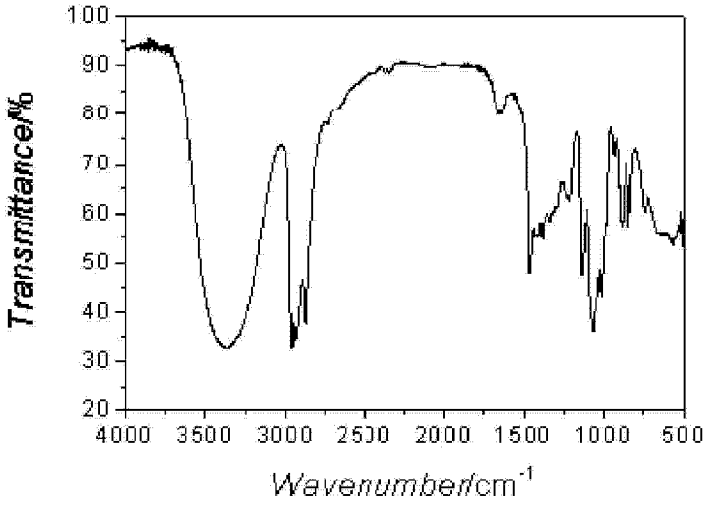
度为 50 % 的过氧化氢水溶液，滴加完毕后在 20 ℃下继续搅拌反应 10 h；然后向反应器中再加入 400 g 水，再在 50 ℃下反应 15 h；在反应器中加入 1 g 二氧化锰，蒸馏回收溶剂甲醇并除去水分，过滤得 292.0 g 纯度为 98.0 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2 戊二醇粗产品；经减压蒸馏，收集 5 mmHg 下 80 ℃左右的馏分，得 278.8 g 纯度为 99.7 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2- 戊二醇产品。

[0031] 实施例 12

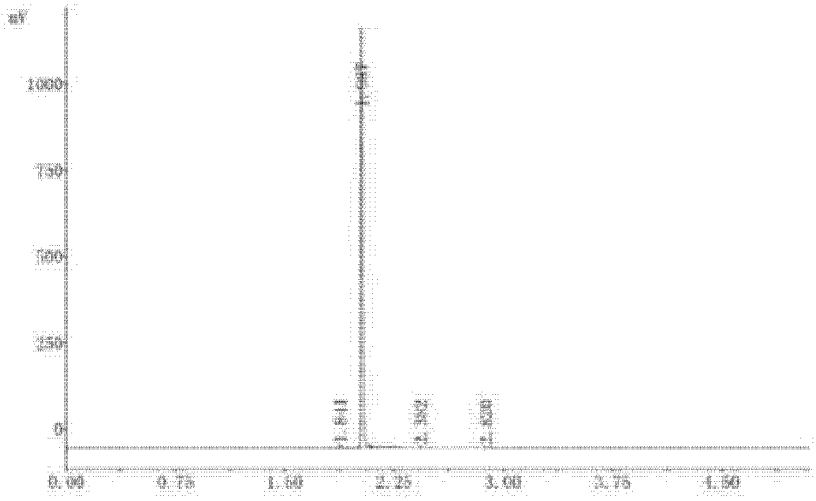
在具有加热、温控和机械搅拌的压热反应器中分别加入 200 mL 甲醇、3.0 g 钛酸四甲酯和 200 g 正戊烯；混合均匀后再加入 300 g 浓度为 30 % 的过氧化氢水溶液，然后在 60 ℃下继续搅拌反应 2 h；加入 400 g 水，再在 60 ℃下反应 10 h；将上述混合液转移到常压蒸馏装置中，蒸馏回收溶剂甲醇并除去水分，过滤得 150.2 g 纯度为 64.0 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2 戊二醇粗产品。

[0032] 实施例 13

在具有加热、温控和机械搅拌的压热反应器中分别加入 200 mL 四氢呋喃、3.0 g 钛酸四甲酯和 200 g 正戊烯；混合均匀后再加入 150 g 浓度为 50 % 的过氧化氢水溶液，然后在 80 ℃下继续搅拌反应 2 h；加入 400 g 水，再在 80 ℃下反应 10 h；将上述混合液转移到常压蒸馏装置中，蒸馏回收溶剂甲醇并除去水分，过滤得 120 g 纯度为 42.0 %（气相色谱，1,2- 丙二醇内标物的内标标准曲线法，）的 1,2 戊二醇粗产品。


Transmittance/%
100
90
80
70
60
50
40
30
20
4000
3500
3000
2500
2000
1500
1000
500
Wavenumber/cm-1

附图 1


mV
1000
750
500
250
0
0.00
0.75
1.50
2.25
3.00
3.75
4.50

附图 2

Bibliographic data: CN104926600 (A) — 2015-09-23

Method for synthesizing 1,2-pentanediol by one-pot method

Page bookmark	CN104926600 (A) - Method for synthesizing 1,2-pentanediol by one-pot method
Inventor(s):	HU XUEYI; CHEN JIAN; FANG YUN +
Applicant(s):	FOSHAN DEPINT ADVANCED MATERIALS CO LTD +
Classification:	- international: *C07C29/03; C07C31/20*
	- cooperative: C07C29/03 → more
Application number:	CN20151239573 20150512 Global Dossier
Priority number(s):	CN20151239573 20150512
Also published as:	CN104926600 (B)

Abstract of CN104926600 (A)

The invention relates to a method for high-yield synthesis of 1,2-pentanediol from n-pentene under low-temperature condition by a one-pot method. According to the method, the reaction temperature is low, so that the risk of leakage caused by high-temperature volatilization of a 1-pentene raw material, and meanwhile, the requirements on pressure resistance of equipment are lowered; by adopting a multistep one-pot method process, process flows are shortened; corrosive, polluting and harmful compounds are not adopted as raw materials during reaction, so that the condition in the traditional process that an organic acid oxygen carrying agent, of which the weight is multiple that of the raw materials, is adopted is avoided; the environment-friendliness of the process is good; and the method has the advantages that the source of raw materials is wide, the production cost is low, and the economical efficiency is high.

DESCRIPTION CN104926600

The invention relates to a method for synthesizing 1,2-pentanediol by high-yield one-pot method from n-pentene as a raw material under low temperature conditions. The reaction temperature of the invention is low, the high temperature volatilization of the 1-pentene raw material is lowered to cause leakage risk, and the pressure resistance requirement of the device is reduced; the multi-step one-pot process is used to shorten the process flow; the reaction is not corroded, The pollution and harmful compounds are used as raw materials, and the organic acid oxygen carrier is used in the traditional process. The process is environmentally friendly; the raw materials of the invention have wide sources, low production cost and good economy.

Method for synthesizing 1,2-pentanediol by one-pot method

Technical field

The invention relates to a one-pot method for synthesizing 1,2-pentanediol, belonging to the field of organic compound synthesis.

Background technique

1

,2

-Pentanediol is an important intermediate for the synthesis of propiconazole fungicides with high activity, low toxicity and broad spectrum of activity (developed by Belgian Janssen Pharmaceutical Group < 1 >, registered in many countries) for the construction of propiconazole molecules. The ketal structure < 2 > . 1,2-Pentanediol also has a moisturizing function, and it is synergistic with other humectants to produce a strong synergistic effect <3>, which can be used as a moisturizing ingredient in cosmetics or personal care products < 3-4 >. 1,2-Pentanediol also has a bacteriostatic < 5 > and can effectively inhibit product deterioration < 6 > in formulations without the addition of other preservatives.

1,2 - The synthesis method of pentanediol is mainly the following two types: (1) epoxidation of n-pentene and subsequent hydrolysis; (2) hydrogenation reduction of furfural or decyl alcohol. The first method mainly has the disadvantages of production safety and pollution. The second method mainly has shortcomings such as low conversion rate of raw materials, many side reactions and difficulty in product purification. The specific conditions are as follows.

Degussa < 7 > discloses a method for synthesizing 1,2-pentanediol from a continuous phase of n-pentene as a raw material, benzene as a solvent, propionic acid as an oxygen-carrying agent and sodium hydroxide as a ring-opening reagent. Ciba-Geigy discloses a continuous production process for the synthesis of 1,2-pentanediol using n-pentene and formic acid as raw materials, hydrogen peroxide as the oxidant and sodium hydroxide as the ring-opening reagent. Zhejiang Liansheng Group < 9 > and Sinopec < 10 > also disclosed similar processes using n-pentene, formic acid and hydrogen peroxide as raw materials. The raw material of the process has a low boiling point (the boiling point of n-pentene is 30.1 ° C), which is easy to cause labor safety and production safety due to volatilization of low-boiling organic compounds; the corrosion of the equipment is strong with formic acid as a raw material, and a small amount of leaked formic acid is used. The operator's skin has a strong corrosive effect, and a large amount of liquid organic waste is treated with 2-10 times of formic acid in the amount of n-pentene material; the by-product of the reaction is many, and the separation and purification of the product is relatively difficult. Dalian Institute of Chemical Industry < 11 > discloses a method for directly preparing 1,2-fatty diol by reacting n-pentene with hydrogen peroxide by phosphotungstic acid, and solves a series of problems caused by excessive formic acid; Under the reaction conditions, low-boiling raw materials volatilize and high pressure resistance requirements for equipment are insufficient. Zhu Yulei, Institute of Coal, Chinese Academy of Sciences < 12 > reported a new green chemical route for the synthesis of 1,2-pentanediol. The manganese dioxide-loaded noble metal ruthenium catalyzed hydrogenation of decyl alcohol to 1,2-pentanediol. The noble metal ruthenium is used in the method, which is expensive; and the supported carrier manganese dioxide is not a carrier material which is particularly suitable for industrial applications. Hou Wei of Tianjin Medical University < 13 > reported that n-pentene is used as raw material, sodium borate is used as oxidant, acetic anhydride is used as reaction solvent and reactant to synthesize monopentyl acetate. The ester compound is further hydrolyzed under alkaline conditions. 1,2-pentanediol. This method avoids highly corrosive compounds such as formic acid. However, excessive acetic anhydride and oxidant lead to difficult post-treatment of the reaction product; stoichiometric reaction of n-pentene with acetic anhydride produces monopentyl acetate, which consumes a large amount of acetic anhydride, increases the cost of raw materials; oxidation reaction forms monoester and hydrolysis The separation of the reaction prolongs the reaction route and increases the operating cost. Severeyns et al. < 14 > catalyze the bis-hydroxylation of n-pentene to synthesize a 1,2-pentanediol diol by the action of NMO and osmium tetroxide catalyst. The ruthenium osmium catalyst used in the method has high toxicity, high price, and is apt to generate toxic gas by heat, so the industrial application value of the route is extremely small.

As early as 1931, Adkins < 15 > reported that copper chromite catalyzed the hydrogenation of sterol to synthesize a mixture of 1,2-pentanediol and 1,5-pentanediol, and added 0.7 g of sterol to 3 g of catalyst at 175 < o > The reaction under C for 11 hours, the total yield of 1,2-pentanediol and 1,5-pentanediol was up to 70%. However, in 1958, Manly used a similar reaction condition of Adkins, and the yield of 1,2-pentanediol was only 4.4%. Tomishige < 17 > at the University of Tsukuba, Japan, published a metal-supported ruthenium and rhodium-catalyzed hydrogenation reduction of decyl alcohol at Chem Comm. As a result, no 1,2-pentanediol was detected in the product, but a selective generation of 1 , 5-pentanediol. Xu Yanjuan of Zhejiang University < 18 > studied Cu-Zn, Cu-Mn, Cu-Cr, Raney Cu, Raney Ni and noble metal supported catalysts Pt/C, Pd/C, Ru/ Al2O3 and other catalytic decyl alcohol selective hydrogenation reduction 1,2-pentanediol, wherein Cu-Zn has the best catalytic performance. The reaction temperature was 150~160 < o > C, the reaction pressure was 7~8MPa, the reaction time was 10h, the conversion of sterol reached 69.2%, the selectivity of 1,2-pentanediol reached 46.2%, and the yield reached 32.0. %. Low conversion of raw materials, high side reactions and low yields of 1,2-pentanediol are still fatal defects in this route. Evonik Industries Group < 19 > discloses a method for the hydrogenation reduction of decyl alcohol to 1,2-pentanediol. Zhuhai Kaimei Technology Co., Ltd. < 20 > discloses the selective hydrogenation of non-precious metal catalysts to 1,2-pentanediol. The method utilizes the deep processing product furfural as raw material of agricultural and sideline products, and greatly solves the problem of limitation of raw materials by n-pentene; the method uses catalytic hydrogenation of non-precious metal catalysts such as copper oxide to solve the problems of expensive and difficult recovery of precious metal catalyst; The method still has problems such as incomplete conversion of raw materials, many side reactions and low yield, which brings new problems such as difficulty in separation, low product purity and non-environmental friendly reaction route; poor performance of catalytic hydrogenation of copper oxide leads to poor performance. The pressure of the hydrogenation reaction is too high, which leads to safety hazards in the production process. Maya Chatterjee < 21 > from the Sendai Chemical Research Center in Sendai, Japan, reported the synthesis of 1,2-pentanediol from tetrahydrofurfuryl alcohol in supercritical carbon dioxide, in which Pd/MCM-41 is the catalyst for 1,2-pentanediol. The highest rate, reaching 77.4%, also contained 12.6% of 1,5-pentanediol and 10% of a mixture of 1-pentanol and 2-pentanol. Although the method improves the yield of 1,2-pentanediol, the supported noble metal is still used as a catalyst, and the operation cost is relatively high; using supercritical carbon dioxide as a solvent, the pressure of the reaction system is also large, and the process is not Suitable for the production of 1,2-pentanediol on an industrial scale.

In addition to the above two methods, there are some sporadic reports on the synthesis of 1,2-pentanediol. Degussa Company < 22 > discloses the nucleophilic reaction of n-butyraldehyde with hydrocyanic acid to form o-hydroxyl-valeronitrile, hydrolyzing o-hydroxy-n-valeronitrile to obtain o-hydroxy-n-pentanol, and then catalyzing a heterogeneous catalyst such as noble metal ruthenium and osmium. Synthesis of 1,2-pentanediol. In the method, the raw material hydrocyanic acid is a highly toxic chemical, which is potentially fatal to the operator; the reaction is carried out in three steps, and the route is long; the first step of the nucleophilic reaction is difficult to control the reaction due to side reactions such as aldol condensation, by-products More, product purification is difficult; the use of precious metals lanthanum and cerium as a catalyst increases production costs. Jiangsu Yangnong Chemical Group < 23 > discloses the epoxidation of 2-chloropentanol in an alkaline solvent. Although the process may obtain 1,2-pentanediol in a high yield, the raw material of 2-chloropentanol is not easily obtained, and the atmospheric distillation of the pentylene oxide in the reaction process has a fatal hidden danger of unsafe operation. Ding Xiaobing of Jiangsu Chemical Pesticide Group reported in "Anhui Chemical" < 24 > Reduction of 1,2-pentanediol by o-hydroxy-n-valeric acid with sodium borohydride. The high cost of sodium borohydride and the inability to obtain hydroxy-p-pentanoic acid make the process difficult to achieve industrial production.

In summary, it has been reported in the literature that the route of synthesizing 1,2 pentanediol by cyclopentene epoxidation reaction and epoxy compound ring-opening reaction has low boiling point and high volatility of raw materials, and liquid waste generated by the process. Many, product separation and post-processing difficulties are not enough; it has been reported in the literature that the route of synthesizing 1,2-pentanediol by hydrogenation reduction of decyl alcohol or furfural solves the problem of raw material source, but the precious metal catalyst still exists. Insufficient conversion rate of raw materials, poor product selectivity and low product purity; other methods reported may have high raw material prices, insufficient source of raw materials, or use of highly toxic chemicals in the reaction process, or long route or reaction Difficulties, or difficulty in product purification.

[References of Background Art]

1. Van Reet, G.; Heeres, J.; Wais, L. Antimicrobial and plant-growth-regulating triazole derivatives. US4160838, 19790103., 1979.

2.

(a) Palecek, J.; Kubelka, V.; Dedek, V.; Mostecky, J. 1-Substituted imidazoles and triazoles. CS215362, 19800522., 1984; (b) Lourens, G. J.; Booysen, M.; Joubert, E. preparation of propiconazole precursor 2-bromomethyl-2-(2,4-dichlorophenyl)-4-propyldioxolane via bromination of 2-(2,4-dichlorophenyl)-2-methyl-4-propyldioxolane. ZA9604976, 19960612., 1997; (c) Darpan, P.; Prasanna, K. A process for the preparation of unsymmetrical triazole fungicides. IN2006MU00697, 20060504., 2008.

3.

Schmaus, G.; Pillai, R. Use of synergistically active 1, 2-alkanediol mixtures as skin moisture-regulating compositions. WO2006069953, 20051221., 2006.

4.

(a) Schrader, K. Alkanediols as skin moisturizers. DE4320744, 19930623., 1995; (b) Serizawa, S. Cosmetic moisturizers containing bisabolol and polyvalent alcohols. JP11060463, 19970819., 1999; (c) Kuroda, A.; Iida, M.; Nakamura, T. Skin-moisturizing cosmetic compositions containing 1,2-pentanediol and isoprene glycol. JP2002047168, 20000801., 2002; (d) Omori, T.; Miyahara, R.; Kanokogi, H.; Nanba, T. Preparation of alkyl polyglycol neopentanoates and their use as moisturizers, cosmetics, and topical use. JP2002302469, 20010330., 2002; (e) Iida, T.; Yamaguchi, F.; Tadenuma, H. Liquid dishwashing detergent compositions and skin-moisturizing method using them. JP2006193732, 20051214., 2006.

5.

Roeding, J.; Lange, S.; Schmaus, G.; Joppe, H.; Pillai, R. Synergistic mixtures of at least one 1, 2 alkanediol such as 1,2-hexanediol and 1,2-octanediol with a further Compound having antimicrobial properties. US20070265352, 20051020., 2007.

6.

(a) Sumino, H.; Asaka, Y. Zinc gluconate and 2-phenoxyethanol or 1,2-pentanediol as antimicrobials for cosmetics. JP2004067626, 20020808., 2004; (b) Merianos, J. J.; Garelick, P.; Lindstrom, S. M. Antimicrobial compositions containing diols. US20050228032, 20040408., 2005; (c) Faergemann, J.; Hedner, T. Antimicrobial compositions including at least three different diols. WO2006057616, 20051129., 2006; (d) Tominaga, H.;

Yamamoto, Y.; Ohashi, Y. Antimicrobial cosmetics or skin preparations containing alkyl glucosides and specific antibacterial agents. JP2010265189, 20090513., 2010.
7.

Siegmeier, R.; Prescher, G.; Maurer, H.; Hering, G. Continuous production of 1,2-pentanediol. DE3442937, 19841124., 1986.
8.

Issler, H.; Maul, R. Continuous process for producing 1,2-alkanediols. US4479021, 19830830., 1984.
9.

Yu Xiaoou A preparation method of 1,2-pentanediol. CN1552684, 2005.
10.

Fu Jiansong; Wang Jiyuan; Sun Chao; Zhang Dongmei; Wu Zhongping A method for preparing 1,2-pentanediol. CN103864575A, 2012.
11.

Gao Shuang; Lu Ying; Zhang Hengyu; Qi Zuwei A preparation method of 1,2-fatty diol. CN1923777, 2005.
12.

Zhang, B.; Zhu, Y.; Ding, G.; Zheng, H.; Li, Y., Selective conversion of furfuryl alcohol to 1,2-pentanediol over a Ru/MnOx catalyst in aqueous phase. Green Chemistry 2012, 14 (12), 3402-3409.
13.

Hou Wei; Liu Jihua Preparation of o-diols by acetic anhydride synthesis. 1996.
14.

Severeyns, A.; De Vos, D. E.; Fiermans, L.; Verpoort, F.; Grobet, P. J.; Jacobs, P. A., A heterogeneous cis-dihydroxylation catalyst with stable, site-isolated osmium -diolate reaction centers. Angewandte Chemie, International Edition 2001, 40 (3), 586-589.
15.

Adkins, H.; Connor, R., Catalytic hydrogenation of organic compounds over copper chromite. Journal of the American Chemical Society 1931, 53 , 1091-5.
16.

Manly, D. G.; Duniop, A. P., Catalytic hydrogenation. I. Kinetics and catalyst composition in the preparation of 2-methylfuran. Journal of Organic Chemistry 1958, 23, 1093-5.
17.

Koso, S.; Furikado, I.; Shimao, A.; Miyazawa, T.; Kunimori, K.; Tomishige, K., Chemoselective hydrogenolysis of tetrahydrofurfuryl alcohol to 1,5-pentanediol. Chemical Communications (Cambridge, United Kingdom) 2009, (15), 2035-2037.

18.

Xu Yanjuan Preparation of 1,2-pentanediol by catalytic hydrogenation of sterol. 2014.

19.

M. Omais; M. Neumann; V. Breme; C. Tess; D. Wolf; P. Claus; M. Lucas; R. Ekter sterol hydrogenolysis into 1,2-pentanediol. CN104016831A.

20.

Cui Jian; Cui Xiaotong, a method for the production of 1,2-pentanediol by one-step hydrogenation of furfural.

21.

Chatterjee, M.; Kawanami, H.; Ishizaka, T.; Sato, M.; Suzuki, T.; Suzuki, A., An attempt to achieve the direct hydrogenolysis of tetrahydrofurfuryl alcohol in supercritical carbon dioxide.
Catalysis Science & Technology 2011, 1 (8), 1466-1471.

22.

Krimmer, H.-P.; Sans, J.; Theis, C.; Thalhammer, F. Process for preparing 1,2-diols from carbonyl compounds via the corresponding cyanohydrins. US20060064905, 20070822., 2006.

23.

Ding Kehong; Cheng Xiaoqing; Zheng Yaqing; Xu Jinlai; Wu Hui; Wang Genlin; Yang Jin; Lu Chunming; Jin Ye A new method for the preparation of 1,2-pentanediol. CN101885668A, 2010.

24.

Ding Xiaobing, the synthesis of 1,2-pentanediol. Anhui Chemical Industry 2002, (05), 22-23+21.

Summary of the invention

[Questions to be solved]

It is an object of the present invention to provide a process for synthesizing 1,2-pentanediol from n-pentene as a raw material under low temperature conditions.

Another object of the present invention is to provide a process for synthesizing 1,2-pentanediol from a raw material which is non-corrosive to equipment.

Another object of the present invention is to provide a more environmentally friendly process for the synthesis of 1,2-pentanediol from 1-pentene.

[Technical solutions]

The method of the present invention comprises the following steps:

1 Adding a certain amount of solvent and catalyst to the reaction kettle and mixing them uniformly;

2 Adding a certain amount of n-pentene to the reaction vessel and mixing uniformly;

3 Keep the temperature of the material in the reactor at 0-40 < o > C, and slowly add a certain amount of hydrogen peroxide under slow stirring conditions for several hours;

4 Adding a certain amount of water or no water to the reaction kettle;

5 Adding a certain amount of manganese dioxide to the reaction kettle;

6 The temperature in the reactor is raised to 30-80 < o > C, and the reaction is for several hours;

7 After the reaction is completed, the solvent in the system is evaporated, and filtered to obtain a crude product of 1,2-pentanediol;

8 The above crude product was distilled under reduced pressure to obtain 1,2-pentanediol having a content of more than 99.5%.

[Beneficial effect]

Compared with the conventional synthesis method of 1,2-pentanediol, the beneficial effects of the present invention are as follows:

1 The reaction temperature provided by the invention is low heat, which reduces the high temperature volatilization of the 1-pentene raw material, leads to the risk of leakage and reduces the pressure resistance requirement for the equipment.

2 The multi-step one-pot reaction process provided by the invention has short route, simple processing and high product yield, and synthesizes 1,2-pentanediol.

3 The process of the invention has no corrosion, no pollution and no harmful compounds, so the synthesis process of the invention has better environmental friendliness.

4 The raw material of the method of the invention has wide source and low cost, short process route, low production cost and good economy.

DRAWINGS

Figure 1 is a Fourier transform infrared spectrum of a potassium bromide salt sheet coating method for 1,2-pentanediol. In the figure, 3361cm < -1 > is the stretching vibration absorption of the associated hydroxyl group; 2958 cm < -1 > , 2931 cm < -1 > and 2872 cm < -1 > are the hydrocarbon stretching vibrations of methyl and methylene groups. Characteristics of absorption.

2 is a gas chromatogram of the product 1,2-pentanediol of Example 2. The gas chromatographic conditions were: OV1701 capillary column, 30 m, 0.32 um, nitrogen as carrier gas, pre-column pressure 0.1 MPa, column temperature 180 < o > C. The area normalized percentage was 99.89%, and the content determined by the internal standard standard curve method using propylene glycol as the internal standard was 99.85%.

Detailed ways

The invention is further illustrated by the following examples, but the invention is not limited to the following examples.

Example 1

In a normal pressure reactor with heating, temperature control, magnetic stirring, constant pressure dropping and reflux device, add 20 mL of methanol and 0.1 g of titanium tetrachloride, mix slowly by slow stirring; under slow stirring conditions 10 g of n-pentene was added dropwise with a constant pressure dropping funnel; under 10 < o > C conditions, 15 g of 30% aqueous hydrogen peroxide solution was slowly added dropwise through a constant pressure separatory funnel at about 30 min, after the addition was completed. Stirring was continued for 6 h at 10 < o > C; then 20 g of water was added to the reactor and reacted for 5 h at 50 < o > C; 0.1 g of manganese dioxide was added to the reactor, and the solvent methanol was distilled off. The water was removed, and 10.3 g of a crude product of 1,2 pentanediol having a purity of 97.3% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard) was filtered.

Example 2

In a normal pressure reactor with heating, temperature control, magnetic stirring, constant pressure dropping and reflux device, add 20 mL of methanol and 0.1 g of tetramethyl titanate respectively, mix slowly by slow stirring; in slow stirring conditions 10 g of n-pentene was added dropwise with a constant pressure dropping funnel; and under a condition of 10 < o > C, 14 g of a 50% aqueous solution of hydrogen peroxide was slowly added dropwise through a constant pressure separatory funnel at about 30 min. After the addition is completed, stirring is continued for 6 h at 10 < o > C; then 30 g of water is added to the reactor, and then reacted at 50 < o > C for 8 h; 0.1 g of manganese dioxide is added to the reactor, and distillation is carried out. The solvent methanol was recovered and the water was removed, and 13.0 g of a crude product of 1,2 pentanediol having a purity of 97.4% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard) was filtered, and distilled under reduced pressure. A fraction of about 80 < o > C at 5 mmHg was collected to obtain 12.1 g of a 1,2-pentanediol product having a purity of 99.6% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard).

Example 3

In a normal pressure reactor with heating, temperature control, magnetic stirring, constant pressure dropping and reflux device, add 20 mL of isopropanol and 1.0 g of tetrabutyl titanate, mix slowly by slow stirring; Under the condition, 10 g of n-pentene was added dropwise by a constant pressure dropping funnel; 40 g of a 10% aqueous solution of hydrogen peroxide was slowly added dropwise to the constant pressure separatory funnel at 10 < o > C for about 30 min. After the addition was completed, stirring was continued for 10 h at 10 < o > C; the temperature of the system was raised to 80 < o > C, and the reaction was continued for 8 h; 0.1 g of manganese dioxide was added to the reactor, and the solvent was distilled to recover isopropanol. The water was removed, and 12.4 g of a crude product of 1,2 pentanediol having a purity of 95.2% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard) was filtered.

Example 4

Add 20 mL of isopropanol and 2.0 g of tetraisopropyl titanate to an atmospheric reactor with heating, temperature control, magnetic stirring, constant pressure dropping and reflux, and mix slowly with slow agitation; 10 g of n-pentene was added dropwise with a constant pressure dropping funnel under stirring; 20 g of a 20% aqueous solution of hydrogen peroxide was slowly added dropwise at 30 ° via a constant pressure separatory funnel at about 10 min. After the addition, the mixture was stirred at 10 < o > C for 10 h; then 20 g of water was added to the reactor, and then reacted at 60 < o > C for 5 h; 0.1 g of manganese dioxide was added to the reactor. The solvent was distilled to recover isopropanol and the water was removed, and filtered to obtain 13.2 g of a crude 1,2 pentanediol having a purity of 96.5% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard).

Example 5

In a normal pressure reactor with heating, temperature control, magnetic stirring, constant pressure dropping and reflux device, add 20 mL of ethanol and 1.5 g of tetrahexyl titanate, mix slowly by slow stirring; in slow stirring conditions 10 g of n-pentene was added dropwise with a constant pressure dropping funnel; 8 g of an aqueous solution of 80% hydrogen peroxide was slowly added dropwise at a pressure of 10 <0>C through a constant pressure separatory funnel at about 30 min. After the addition is completed, stirring is continued for 2 h at 40 < o > C; then 20 g of water is added to the reactor, and then reacted at 70 < o > C for 4 h; 0.1 g of manganese dioxide is added to the reactor, and distillation is carried out. The solvent ethanol was recovered and the water was removed, and 12.5 g of a crude product of 1,2 pentanediol having a purity of 95.3% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard) was filtered, and distilled under reduced pressure. A fraction of about 80 < o > C at

5 mmHg was collected to obtain 11.2 g of a 1,2-pentanediol product having a purity of 99.5% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard).

Example 6

In a normal pressure reactor with heating, temperature control, magnetic stirring, constant pressure dropping and reflux device, add 30 mL of acetone and 1.0 g of tetraisobutyl titanate respectively, mix slowly by slow stirring; in slow stirring conditions 10 g of n-pentene was added dropwise with a constant pressure dropping funnel; 20 g of a 30% aqueous solution of hydrogen peroxide was slowly added dropwise at a pressure of 10 <0>C through a constant pressure separatory funnel at 30 min. After the addition is completed, stirring is continued for 4 h at 20 < o > C; then 10 g of water is added to the reactor, and then reacted at 40 < o > C for 20 h; 0.1 g of manganese dioxide is added to the reactor, and distillation is carried out. The solvent acetone was recovered and the water was removed, and 12.7 g of a crude 1,2 pentanediol having a purity of 95.9% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard) was filtered.

Example 7

In a normal pressure reactor with heating, temperature control, magnetic stirring, constant pressure dropping and reflux device, add 20 mL of methyl ethyl ketone and 0.3 g of titanium tetrachloride, mix slowly by slow stirring; under slow stirring conditions 10 g of n-pentene was added dropwise by a constant pressure dropping funnel; 12 g of a 30% aqueous solution of hydrogen peroxide was slowly added dropwise at a pressure of 10 <0>C through a constant pressure separatory funnel at 30 min. After completion, stir at 30 < o > C for 2 h; then add 10 g of water to the reactor, and then react at 50 < o > C for 16 h; add 0.1 g of manganese dioxide to the reactor, and recycle it. The solvent was subjected to methyl ethyl ketone and water was removed, and 11.2 g of a crude product of 1,2 pentanediol having a purity of 98.1% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard) was filtered.

Example 8

In a normal pressure reactor with heating, temperature control, magnetic stirring, constant pressure dropping and reflux device, add 20 mL of tetrahydrofuran and 0.2 g of tetramethyl titanate respectively, mix slowly by slow stirring; under slow stirring conditions 10 g of n-pentene was added dropwise with a constant pressure dropping funnel; under a condition of 10 < o > C, 15 g of a 30% aqueous solution of hydrogen peroxide was slowly added dropwise through a constant pressure separatory funnel at about 30 min. After the addition is completed, stirring is continued for 1 h at 40 < o > C; then 20 g of water is added to the reactor, and then reacted at 50 < o > C for 4 h; 0.1 g of manganese dioxide is added to the reactor, and distillation is carried out. The solvent tetrahydrofuran was recovered and the water was removed, and 14.5 g of crude 1,2 pentanediol having a purity of 98.3% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard) was filtered, and distilled under reduced pressure. A fraction of about 80 < o > C at 5 mmHg was collected to obtain 13.5 g of a 1,2-pentanediol product having a purity of 99.9% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard).

Example 9

In a normal pressure reactor with heating, temperature control, magnetic stirring, constant pressure dropping and reflux device, add 20 mL of diethyl ketone and 0.3 g of tetraethyl titanate respectively, mix slowly by slow stirring; 10 g of n-pentene was added dropwise with a constant pressure dropping funnel under stirring; 10 g of a 50% aqueous solution of hydrogen peroxide was slowly added dropwise to the constant pressure separatory funnel at 10 < o > C for about 30 min. After the addition, the mixture was stirred at 20 < o > C for 10 h; then 20 g of water was added to the reactor, and then reacted at 50 < o > C for 4 h; 0.1 g of manganese dioxide was added to the reactor. Distilling solvent tetrahydrofuran and removing water, and filtering 13.7 g of crude 1,2 pentanediol having a purity of 98.0%

(gas chromatography, internal standard curve method of 1,2-propanediol internal standard); Distillation, collecting fractions of about 80 < o > C at 5 mmHg, yielding 12.4 g of 1,2-pentanediol with a purity of 99.7% (gas chromatography, internal standard curve method for 1,2-propanediol internal standard) product.

Example 10

Add 200 mL of anhydrous methanol and 3.0 g of tetramethyl titanate to a normal pressure reactor with heating, temperature control, magnetic stirring, constant pressure dropping and reflux, and mix slowly by slow stirring; 200 g of n-pentene was added dropwise with a constant pressure dropping funnel under stirring; 300 g of a 30% aqueous solution of hydrogen peroxide was slowly added dropwise to the constant pressure separatory funnel at 10 < o > C for about 30 min. After the addition, the mixture was stirred at 10 < o > C for 10 h; then 200 g of water was added to the reactor, and then reacted at 50 < o > C for 14 h; 1 g of manganese dioxide was added to the reactor. Distilling solvent methanol and removing water, and filtering to obtain 290.2 g of a crude product of 1,2 pentanediol having a purity of 98.1% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard); Distillation, collecting fractions of about 80 < o > C at 5 mmHg, yielding 281.0 g of 1,2-pentanediol with a purity of 99.7% (gas chromatography, internal standard curve method for 1,2-propanediol internal standard) product.

Example 11

Add 200 mL of tetrahydrofuran and 3.0 g of tetramethyl titanate to an atmospheric reactor with heating, temperature control, magnetic stirring, constant pressure dropping and reflux, and mix slowly by slow stirring; 200 g of n-pentene was added dropwise with a constant pressure dropping funnel; 150 g of a 50% aqueous solution of hydrogen peroxide was slowly added dropwise at a pressure of 10 <0>C through a constant pressure separatory funnel at 30 min. After the addition is completed, the reaction is further stirred at 20 < o > C for 10 h; then 400 g of water is added to the reactor, and then reacted at 50 < o > C for 15 h; 1 g of manganese dioxide is added to the reactor. The solvent methanol was distilled off and the water was removed, and 292.0 g of a crude 1,2-pentanediol having a purity of 98.0% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard) was obtained by filtration; Collecting a fraction of about 80 < o > C at 5 mmHg to obtain 278.8 g of a 1,2-pentanediol product having a purity of 99.7% (gas chromatography, internal standard curve method for 1,2-propanediol internal standard) .

Example 12

200 mL of methanol, 3.0 g of tetramethyl titanate and 200 g of n-pentene were added to the autoclave with heating, temperature control and mechanical stirring; after mixing, 300 g of hydrogen peroxide at a concentration of 30% was added. The aqueous solution is then stirred at 60 < o > C for 2 h; 400 g of water is added, and then reacted at 60 < o > C for 10 h; the mixture is transferred to an atmospheric distillation unit, and the solvent methanol is distilled off. The water was removed, and 150.2 g of a crude 1,2 pentanediol having a purity of 64.0% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard) was filtered.

Example 13

200 mL of tetrahydrofuran, 3.0 g of tetramethyl titanate and 200 g of n-pentene were added to the autoclave with heating, temperature control and mechanical stirring; after mixing, 150 g of 50% hydrogen peroxide was added. The aqueous solution is then stirred at 80 < o > C for 2 h; 400 g of water is added, and then reacted at 80 < o > C for 10 h; the mixture is transferred to an atmospheric distillation unit, and the solvent methanol is distilled off. The water was removed, and 120 g of a crude 1,2 pentanediol having a purity of 42.0% (gas chromatography, internal standard curve method of 1,2-propanediol internal standard) was filtered.

CLAIMS CN104926600

1.

The invention relates to a method for synthesizing 1,2-pentanediol by one-pot method, which is characterized in that 1-pentene, hydrogen peroxide and a catalyst are heated in a solvent to a certain temperature for a certain reaction.

2.

The hydrogen peroxide according to claim 1, characterized in that the hydrogen peroxide concentration is from 10% to 80% (w/w).

3.

The mass ratio of 1-pentene to hydrogen peroxide according to claim 1 or 2, characterized in that the ratio is 1:1 to 2.

4.

The catalyst according to claim 1, wherein the catalyst is titanium tetrachloride or a titanate having the formula of Ti(OR) 4 , and R is a C 1 - C 10 alkyl group.

5.

The catalyst according to claim 1 or 4, which is characterized in that the amount is from 0.1% to 10% (w/w) based on the mass of the raw material 1-pentene.

6.

The reaction temperature of claim 1 is characterized by a range of 10 < o > C-100 < o > C.

7.

The reaction temperature of claim 1 or claim 6 which is characterized by a constant temperature reaction.

8.

The reaction temperature of claim 1 or claim 6 characterized by a temperature programmed reaction.

9.

The reaction time described in claim 1 is from 1 h to 24 h.

10.

The solvent according to claim 1, wherein the solvent is one or a mixture of methanol, ethanol, isopropanol, n-butanol, acetone, methyl ethyl ketone, and dibutyl ketone.

(19)日本国特許庁(JP)　(12)公開特許公報(A)　(11)特許出願公開番号
特開2008-7463
(P2008-7463A)
(43)公開日　平成20年1月17日(2008.1.17)

(51)Int.Cl.		F I			テーマコード（参考）
C07C 29/128	(2006.01)	C07C	29/128		4H006
C07C 31/20	(2006.01)	C07C	31/20	Z	4H039
C07B 61/00	(2006.01)	C07B	61/00	300	

審査請求 未請求　請求項の数 3　OL　（全 11 頁）

(21)出願番号　特願2006-179791 (P2006-179791)
(22)出願日　平成18年6月29日 (2006.6.29)

(71)出願人　000205638
大阪有機化学工業株式会社
大阪府大阪市中央区安土町１丁目７番２０号
(74)代理人　100095832
弁理士　細田　芳徳
(72)発明者　山本　達郎
大阪府柏原市片山町１８番８号　大阪有機化学工業株式会社内
(72)発明者　吉丸　俊彦
大阪府柏原市片山町１８番８号　大阪有機化学工業株式会社内

Fターム(参考)　4H006 AA02 AC41 BA52 BA66 BB14 BC34 BE32 FC50 FE11 FG29
4H039 CA60 CA63 CE10 CH40

(54)【発明の名称】１，２－アルカンジオールの製造法

(57)【要約】
【課題】低臭気の１，２－アルカンジオールを高純度で容易かつ安全に製造しうる方法を提供すること。
【解決手段】ギ酸の存在下で、オレフィンと過酸化水素とを反応させ、得られた反応混合物とアルコールとを反応させることを特徴とする１，２－アルカンジオールの製造法。
【選択図】なし

【特許請求の範囲】
【請求項１】
ギ酸の存在下で、オレフィンと過酸化水素とを反応させ、得られた反応混合物とアルコールとを反応させることを特徴とする１，２－アルカンジオールの製造法。
【請求項２】
オレフィンが、式：

$$R^1R^2C=CR^3R^4 \qquad (I)$$

（式中、R^1、R^2、R^3およびR^4は、それぞれ独立して、水素原子、置換基を有していてもよいアルキル基または置換基を有していてもよいアリール基を示すが、R^1、R^2、R^3およびR^4のうちの２つの基が一緒になって環を形成していてもよい）
で表されるオレフィンである請求項１記載の製造法。
【請求項３】
過酸化水素の量が、オレフィンの二重結合に対して、０．５～１当量である請求項１または２記載の製造法。
【発明の詳細な説明】
【技術分野】
【０００１】
本発明は、１，２－アルカンジオールの製造法に関する。さらに詳しくは、潤滑油、化粧品、香料、医薬品などの原料として有用な１，２－アルカンジオールの製造法に関する。
【背景技術】
【０００２】
１，２－アルカンジオールの製造法として、ギ酸または酢酸の存在下で、オレフィンと過酸化水素とを反応させ、生成したエポキシドを加水分解させる方法が知られている（例えば、非特許文献１および２参照）。しかし、この方法には、反応終了後に、溶媒の除去、水酸化ナトリウム水溶液との反応、および塩酸による中和などの多段階の操作を必要とするため、製造効率の点で欠点がある。
【０００３】
また、１，２－ジオールの製造法として、過酢酸を酸化剤として用い、オレフィンからエポキシドを経由して１，２－ジオールを製造する方法が知られている（例えば、特許文献１参照）。しかし、この方法には、エポキシドとジオールによる副生成物が生じるという欠点がある。
【０００４】
一方、エポキシドとジオールによる副生成物が生じることを回避しうる１，２－ジオールの製造法として、触媒としてタングステン錯体の存在下で、過酸化水素水とオレフィンとを反応させて１，２－ジオールを製造する方法が知られている（例えば、非特許文献３～５参照）。しかし、この方法には、生成したジオールがさらに酸化されるため、ケトンやカルボン酸を副生するという欠点がある。
【０００５】
反応条件を工夫することにより、１，２－ジオールの選択性を高める方法も提案されている（例えば、特許文献２および非特許文献６参照）。しかし、この方法には、毒性の高いベンゼンを溶媒として用いるため、作業環境における安全性に欠点があり、さらに生成物を単離するために３０％硫酸水溶液で抽出し、塩基で中和するという煩雑な操作を必要とするため、製造効率の点でも欠点がある。
【０００６】
また、他の１，２－ジオールの製造法として、濾過により容易に分離することができる固体触媒を用いてオレフィンと過酸化水素水とを反応させる方法が提案されている（例えば、非特許文献７～９参照）。しかし、この方法には、反応操作上での改善が認められるが、目的化合物とともにエポキシドやケトンを多量に生じるため、１，２－ジオールの収率および選択性が低いという欠点がある。

【０００７】
他の１，２－ジオール類の製造法として、スルホン酸基を有する高分子化合物の存在下に、オレフィン類と過酸化水素とを反応させることを特徴とする１，２－ジオール類の製造法が提案されている（例えば、特許文献３参照）。しかし、この方法には、反応時間をはじめ、効率面や安全性の面で欠点がある。
【０００８】
また、従来の１，２－アルカンジオールには、微量の不純物が含まれていることなどにより臭気が強いものが多いため、香料、化粧品などの用途に原料として用いられる１，２－アルカンジオールには、低臭気であることが求められている。
【０００９】
【特許文献１】特開平４－４１４４９号公報
【特許文献２】欧州特許第１４６３７４号明細書
【特許文献３】特開２００３－２１２８０４号公報
【非特許文献１】オーガニック・シンセシス(Organic Syntheses), Coll. ３巻、２１７－２１９頁（１９５５年）
【非特許文献２】ジャーナル・オブ・アメリカン・ケミカル・ソサイエティー(J. Am. Chem. Soc.）６７巻、１７８６－１７８８頁（１９４５年）
【非特許文献３】ジャーナル・オブ・ケミカル・ソサイエティー(J. Chem. Soc.)２９８８－３０００頁（１９４９年）
【非特許文献４】ジャーナル・オブ・アメリカン・オイル・ケミカル・ソサイエティー(J. Am. Oil Chem. Soc.）４４巻、３１６－３２０頁（１９６７年）
【非特許文献５】テトラヘドロン・レターズ(Tetrahedron Lett.）２９巻、８２３－８２６頁（１９８８年）
【非特許文献６】シンセシス(Synthesis）２９５－２９７頁(１９８９年)
【非特許文献７】ジャーナル・オブ・キャタリスト(J. Cat.）１４５巻、１５１－１５８頁（１９９４年）
【非特許文献８】ケミカル・コミュニケーションズ(Chem. Commun.）３２５－３２６頁(１９９８年)
【非特許文献９】ジャーナル・オブ・キャタリスト(J. Cat.）１８９巻、４０－５１頁（２０００年）
【発明の開示】
【発明が解決しようとする課題】
【００１０】
本発明は、前記従来技術に鑑みてなされたものであり、低臭気の１，２－アルカンジオールを高純度で容易かつ安全に製造しうる方法を提供することを課題とする。
【課題を解決するための手段】
【００１１】
本発明者らは、前記従来技術に鑑みて鋭意研究を重ねた結果、ギ酸の存在下で、オレフィンと過酸化水素とを反応させ、得られた反応混合物とアルコールとを反応させた場合には、１，２－アルカンジオールが高純度で容易に得られ、しかも得られた１，２－アルカンジオールは、臭気をほとんど生じさせないという驚くべき効果が奏されることを見出した。
【００１２】
本発明は、かかる知見に基づいて完成されたものであり、ギ酸の存在下で、オレフィンと過酸化水素とを反応させ、得られた反応混合物とアルコールとを反応させることを特徴とする１，２－アルカンジオールの製造法に関する。
【発明の効果】
【００１３】
本発明の製造法によれば、低臭気の１，２－アルカンジオールを高純度で容易かつ安全に製造することができるという効果が奏される。

【発明を実施するための最良の形態】

【００１４】

本発明においては、まず、ギ酸の存在下で、オレフィンと過酸化水素とを反応させる。

【００１５】

オレフィンとしては、種々のものを広範囲で用いることができる。代表的なオレフィンとしては、例えば、式（Ｉ）：

$$R^1R^2C=CR^3R^4 \qquad (I)$$

（式中、R^1、R^2、R^3およびR^4は、それぞれ独立して、水素原子、置換基を有していてもよいアルキル基または置換基を有していてもよいアリール基を示すが、R^1、R^2、R^3およびR^4のうちの２つの基が一緒になって環を形成していてもよい）

で表されるオレフィンが挙げられ、これらは、それぞれ単独でまたは２種以上を混合して用いることができる。

【００１６】

アルキル基としては、炭素数１～３５のアルキル基が好ましく、炭素数が１～２１のアルキル基がより好ましい。アルキル基が有していてもよい置換基としては、例えば、水酸基、炭素数１～２２のアルコキシ基などが挙げられる。

【００１７】

アリール基としては、炭素数６～３５のアリール基が好ましく、炭素数６～２２のアリール基がより好ましい。アリール基が有していてもよい置換基としては、例えば、水酸基、フェニル基、炭素数１～２２のアルコキシ基などが挙げられる。

【００１８】

なお、R^1、R^2、R^3およびR^4のうちの２つの基が一緒になって環を形成していてもよい。かかる環の例としては、炭素数３～３５のシクロアルキル基などが挙げられ、また、これらの環は他の環と縮合環、非縮合環等の多環を形成していてもよい。更にはR^1、R^2、R^3及びR^4の何れか１つから水素原子を取り除いた残基が２価の原子及び／又は２価の官能基を介して互いに結合して、それらが結合している炭素原子と一緒になって環を形成していてもよいが、この場合の２価の原子としては、酸素原子、窒素原子、硫黄原子等が、また２価の官能基としてはシリレン基、エチレンジオキシ基、アリーレンジオキシ基、カルボニル基、スルホキシド基、スルホン基等が例示される。

【００１９】

式（Ｉ）で表されるオレフィンの具体例としては、１－ヘキセン、２，３－ジメチル－２－ブテン、２－メチル－２－ペンテン、２－ヘプテン、シクロペンテン、シクロヘキセン、１－メチルシクロヘキセン、シクロオクテン、シクロドデセン、１－ウンデセン、１－ドデセン、２－シクロヘキセン－１－オール、１－エチル－１－シクロペンテン、けい皮アルコール、２－メチル－３－フェニル－２－プロペン－１－オール、４－（１－プロペニル）－１，２－ジメトキシベンゼンなどが挙げられる。

【００２０】

過酸化水素は、通常、水溶液として用いられる。過酸化水素水溶液における過酸化水素の濃度には特に制限がないが、通常、好ましくは１～８０％、より好ましくは３～６０％である。

【００２１】

過酸化水素の量は、オレフィンの二重結合に対して、当量以下が好ましい。通常、過酸化水素の量は、過酸化水素がオレフィンに対して過剰な場合は、反応後に過剰の過酸化物を処理する必要があり、場合によっては処理に用いたものの洗浄分離や濾過による除去工程も必要となる点、また過酸化物の処理剤により臭気成分が発生したり副反応物が生成したりする（通常、チオ硫酸ナトリウムなどの場合は、その硫黄原からの臭気が発生しやすく、酸化マンガンなどの触媒の場合には、ジオールの酸化などの副反応や、発熱による臭気成分や不純物が生じやすい）場合が多い点、および過酸化物過剰による危険性の増加による安全面の観点から、オレフィンの二重結合に対して、好ましくは０．５～１当量、より好ましくは０．７～０．９５当量である。

【００２２】
ギ酸の量は、オレフィンの酸化をより温和な条件で安全に進行させるため、反応系内において過ギ酸を生成させること、およびオレフィンから中間体として生成するエポキシ体とジオールとの副反応を抑制するため、エポキシ中間体をギ酸と反応させて開環させる観点から、通常、オレフィンの二重結合に対して、好ましくは１～３０当量、より好ましくは１～１５当量、生産効率の観点から、さらに好ましくは１～５当量である。
【００２３】
オレフィンと過酸化水素との反応は、例えば、オレフィンとギ酸とを混合した後、得られた混合物を撹拌しながら、該混合物に過酸化水素を添加する方法により、容易に行うことができる。
【００２４】
オレフィンと過酸化水素との反応温度は、通常、２０～１００℃、好ましくは３０～６０℃である。
【００２５】
なお、オレフィンと過酸化水素との反応の際には、溶媒を用いてもよい。溶媒としては、例えば、エーテル類、エステル類、ニトリル類、アミド類、スルホキシド類、炭化水素化合物などが挙げられる。溶媒の量は、特に制限されないが、一般的には、オレフィン１００容量部あたり、好ましくは１０００容量部以下、より好ましくは３００容量部以下である。
【００２６】
オレフィンと過酸化水素との反応の終点は、例えば、ガスクロマトグラフィーによる分析及び、過酸化物の滴定による過酸の消失確認などにより、容易に確認することができる。
【００２７】
かくしてオレフィンと過酸化水素との反応により、生成したエステルを含有する反応混合物が得られる。得られた反応混合物は、必要により、濃縮してもよい。
【００２８】
次に、得られた反応混合物とアルコールとを反応させる。かかる反応は、具体的には、得られた反応混合物に含まれている生成したエステルとアルコールとのエステル交換反応である。
【００２９】
アルコールとしては、例えば、炭素数１～４の直鎖または分岐鎖を有する１価アルコールが挙げられる。かかる１価アルコールの具体例としては、メタノール、エタノール、イソプロパノールなどが挙げられるが、本発明は、かかる例示のみに限定されるものではない。
【００３０】
アルコールの量は、反応混合物に含まれるエステル部位と残留しているギ酸を完全にギ酸エステル化する観点から、反応混合物に含まれている生成したエステルのエステル部と残留しているギ酸の合計に対して、好ましくは１～１０当量、より好ましくは２～１０当量、さらに好ましくは３～５当量である。
【００３１】
なお、得られた反応混合物とアルコールとの反応の際には、触媒を用いることができる。触媒としては、一般にエステル交換反応の際に使用されるものであればよい。触媒の例としては、硫酸、パラトルエンスルホン酸などのスルホニル化合物、スルホン酸基を有する高分子化合物などの酸触媒、リチウム化合物、チタン化合物、ナトリウム化合物、アルミニウム化合物、スズ化合物、亜鉛化合物などの金属触媒などが挙げられるが、本発明は、かかる例示のみに限定されるものではない。
【００３２】
触媒の量は、特に制限されないが、一般的には、反応混合物１００重量部あたり、好ましくは０．０１～５重量部、より好ましくは０．０５～１重量部である。

【００３３】
また、オレフィンと過酸化水素との反応は、通常溶媒を用いずに行うこともできるが、溶媒を用いることも可能である。溶媒としては、一般に酸化反応やエステル交換反応の際に使用されるものを用いることができる。溶媒の例としては、エーテル類、ニトリル類、アミド類、スルホキシド類、炭化水素化合物などが挙げられる。溶媒の量は、特に制限されないが、一般的には、オレフィン１００容量部あたり、好ましくは５０～１０００容量部、より好ましくは１００～３００容量部である。
【００３４】
得られた反応混合物とアルコールとの反応は、例えば、反応混合物に、アルコールおよび必要により触媒を添加し、混合することによって行うことができる。
【００３５】
反応温度は、通常、２０～８０℃の範囲内であることが好ましく、生成するギ酸エステルを選択的に反応系外へ出して、平衡反応を進行させる観点から、用いられるアルコールの沸点以下および生成するギ酸エステルの沸点以上であることがより好ましい。反応の際には、生成するギ酸エステルを回収することが好ましいが、特に、アルコールとしてメタノールを用いた場合には、他のギ酸エステルに比べて沸点が低いギ酸メチルが生成するため、より温和な条件下で反応混合物とアルコールとの反応を行うことができ、またメタノールとギ酸メチルは共沸せず、ギ酸メチルのみが揮発するため、ギ酸メチルを容易に反応系外に除去することができる。
【００３６】
反応混合物とアルコールとの反応は、通常溶媒を用いずに行うこともできるが、溶媒を用いることも可能である。溶媒としては、エステル交換反応の際に使用されるものを用いることができる。溶媒の例としては、エーテル類、ニトリル類、アミド類、スルホキシド類、炭化水素化合物などが挙げられる。
【００３７】
反応混合物とアルコールとの反応の終点は、例えば、ガスクロマトグラフィーによる分析などにより容易に確認することができる。
【００３８】
かくして、目的化合物である１，２－アルカンジオールが得られる。なお、反応の際に触媒を用いた場合には、生成した１，２－アルカンジオールを含有する反応混合物にアルカリを添加することにより、触媒を中和することが好ましい。
【００３９】
なお、１，２－アルカンジオールを含有する反応混合物に含まれているギ酸エステル、ギ酸およびアルコールは、反応混合物を濃縮することにより回収し、再利用することができる。得られた１，２－アルカンジオールは、例えば、蒸留などによって精製してもよい。また、１，２－アルカンジオールが、水と混合した際に少なくとも水層への移行が可能な程度の水溶性を示す場合には、１，２－アルカンジオールを、ノルマルヘキサン、シクロヘキサン、２－メチルヘキサン等の疎水性溶剤及び水と混合することにより、臭気成分は有機層に、１，２－アルカンジオールは水層にそれぞれ移行し、より臭気の低減された１，２－アルカンジオールを水層に抽出することができる。
【００４０】
本発明の製造法は、毒性の高いベンゼンなどの溶媒を必要しないので、作業環境における安全性に優れている。また、本発明の製造法によれば、１，２－アルカンジオールを高純度でかつ高収率で容易に得ることができる。さらに、本発明の製造方法で得られた１，２－アルカンジオールは、ほとんど臭気を発しないので、潤滑油、化粧品、香料、医薬品などの原料として好適に使用することができる。
【実施例】
【００４１】
次に、本発明を実施例に基づいてさらに詳細に説明するが、本発明はかかる実施例のみに限定されるものではない。

7/31/2019, EAST Version: 4.1.2.6

【００４２】
実施例１
（１）オレフィンと過酸化水素との反応
１－ヘキセン１００ｇおよびギ酸（純度９５％）２３７．５ｇを１リットル容のビーカー内で混合し、得られた混合物を攪拌しながら加熱し、３０℃に到達した時点で３５％過酸化水素水溶液９８．２ｇを１０時間かけてゆっくりと前記混合物に添加した。反応により発熱するので、混合物の温度が３０～４０℃となるように外部から冷却した。
【００４３】
過酸化水素水溶液の添加終了後、混合物の温度を３５℃で２時間保持することにより、反応を終了した。反応の終点は、過酸化物量の滴定(ヨウ化カリウム滴定法)により確認した。得られた反応混合物を過剰分のギ酸の留出が少なくなるまで(ギ酸もギ酸エステルとなるため完全に濃縮する必要は無い)となるまで濃縮し、濃縮された反応混合物を次のアルコールとの反応に供した。
【００４４】
（２）反応混合物とアルコールとの反応
前記（１）で得られた濃縮された反応混合物１７６．７ｇ、メタノール１７０ｇおよびパラトルエンスルホン酸一水和物０．０９ｇを１リットル容のビーカー内で混合し、生成するギ酸メチルをビーカーから取り出しながら５５℃で７時間攪拌した。その後、ビーカー内に３０％水酸化ナトリウム水溶液０．０８ｇを添加し、触媒（パラトルエンスルホン酸一水和物）の中和を行った後、メタノールを減圧下で留去し、１，２－ヘキサンジオール１２０．７ｇを得た。その結果、１，２－ヘキサンジオールの収率は９２％であり、１，２－ヘキサンジオールの純度は、９２％であった。
【００４５】
次に、得られた１，２－ヘキサンジオールを蒸留により精製したところ、収率７６％、初留をさらに再蒸留することにより、蒸留収率９４％で、９８％以上の純度を有する１，２－ヘキサンジオールが得られた。
【００４６】
実施例２
実施例１と同様にして１，２－ヘキサンジオールを製造し、蒸留による精製まで行った。得られた１，２－ヘキサンジオール５ｇ（純度９８％）、ノルマルヘキサン１０ｇおよび水１０ｇを２５℃で混合撹拌した。ヘキサン層を分離し、水層を濃縮し、臭気成分が除去された、１，２－ヘキサンジオール４．９ｇ（ノルマルヘキサンおよび水と混合した１，２－ヘキサンジオールに対する収率９８％、純度９９％）を得た。
【００４７】
実施例３～９
１－ヘキセンの代わりに表１に示すオレフィンを使用し、ギ酸、過酸化水素水溶液等を表１に示す量で使用した以外は、実施例１と同様にして、１，２－アルカンジオールを得た。得られた１，２－アルカンジオール及びその収率、純度を表１に示す。
【００４８】

7/31/2019, EAST Version: 4.1.2.6

【表１】

	（１）				過酸化水素水溶液の添加時間(hr)	（２）			得られた1,2-アルカンジオール			
	オレフィン		ギ酸(g)	35%過酸化水素水溶液(g)		反応混合物(g)	メタノール(g)	パラトルエンスルホン酸一水和物(g)	種類	収量(g)	収率(%)	純度(%)
	種類	使用量(g)										
実施例1	1-ヘキセン	100.0	237.5 (4.1)	98.2 (0.85)	10	176.7	170	0.09	1,2-ヘキサンジオール	120.7	92	92
実施例3	4-メチル-1-ペンテン	100.0	259.1 (4.5)	98.2 (0.85)	10	170.7	170	0.09	4-メチル-1,2-ペンタンジオール	121.9	91.9	90
実施例4	シクロヘキセン	10.0	26.5 (4.5)	10.1 (0.85)	10	175.0	17	0.009	1,2-シクロヘキサンジオール	12.0	93.9	94
実施例5	1-ドデセン	10.0	13.0 (4.5)	4.9 (0.85)	10	132.5	9	0.009	1,2-ドデカンジオール	10.2	88.8	89
実施例6	1-オクテン	15.1	29.2 (4.5)	11.1 (0.85)	12	20.1	29	0.1	1,2-オクタンジオール	15.1	90.7	99.1
実施例7	4,4-ジメチル-1-ペンテン	5.1	12.6 (5.0)	4.3 (0.85)	10	6.6	13	0.01	4,4-ジメチル-1,2-ペンタンジオール	4.7	82.6	99.5
実施例8	4-メチル-1-ヘキセン	7.0	17.2 (5.0)	5.9 (0.85)	10	9.2	18	0.02	4-メチル-1,2-ヘキサンジオール	6.0	75.6	98.7
実施例9	5-メチル-1-ヘキセン	6.6	42.6 (13.1)	5.6 (0.85)	10	9.6	9	0.02	5-メチル-1,2-ヘキサンジオール	6.2	97.5	99.6

注）ギ酸及び過酸化水素水溶液の使用量に記載の"()"内の数値は、ギ酸又は過酸化水素のオレフィンの二重結合に対する当量数を示す。

【００４９】

7/31/2019, EAST Version: 4.1.2.6

比較例１

シクロヘキセン４１ｇをガラスのフラスコに仕込み、攪拌しながら３０℃に昇温する。３０℃に達したところで３０％の過酢酸をモル比で１．２倍になるように２時間かけてゆっくり添加した。反応により発熱するので混合物の温度が３０℃程度になるように、外部から冷却した。過酢酸の添加終了後、混合物の温度を３０℃で１時間保持することにより、反応を完結させた。反応粗液を、ガスクロマトグラフィーにより分析した結果、シクロヘキセンオキシドが収率の９３％で得られていることを確認した。

【００５０】

水５０ｇをガラス製のフラスコに仕込み、撹拌しながら加熱し、７０℃に到達した時点で、前記で得られたシクロヘキセンオキシドおよび酢酸を含む溶液２１．６ｇを２時間かけてゆっくり添加した。溶液の添加終了後、すぐに反応粗液をガスクロマトグラフィーにより分析した結果、シクロヘキサン－１，２－ジオールが、シクロヘキセンオキシド基準では９４％の収率で、シクロヘキセン基準では８７．４％の収率で得られていることを確認した。

【００５１】

比較例２

Ｎａｆｉｏｎ－ＮＲ５０（ＤｕＰｏｎｔ社製）５００ｍｇ(オレフィンに対するスルホン酸基の当量は０．０４に相当）、３０％過酸化水素水溶液２．２３ｇ（１９．６ｍｍｏｌ）および１－ヘキセン８２５ｍｇ（９．８１ｍｍｏｌ）を混合し、７０℃で２４時間撹拌した。ガスクロマトグラフィーで反応溶液を分析した結果、ほとんど反応の進行は認められなかった。

【００５２】

反応溶液からＮａｆｉｏｎ－ＮＲ５０を濾過して取り除き、溶液中に過剰に存在する過酸化水素を分解するため二酸化マンガン１０ｍｇ（０．１１５ｍｍｏｌ）を加えた。反応溶液を濾過した後、ロータリーエバポレーターを用いて水等を留去したところ、１，２－ヘキサンジオールを濃縮混合物として得た（収率：５％未満）。

【００５３】

比較例３

実施例１の（１）オレフィンと過酸化水素との反応と同様にして得られた、濃縮された反応混合物１７６．７ｇに、３０％水酸化ナトリウム水溶液２３９．３ｇを、２０～３５℃で滴下した。反応の終点は、ガスクロマトグラフィーによる分析により確認した。得られた反応混合物に、抽出溶剤のメチルターシャリーブチルエーテル１４２．４ｇを加え、撹拌後、静置しギ酸カリウムが含まれる水層を分離した。有機層に、２０％食塩水７１．２ｇを加え洗浄し、撹拌、静置後水層を分離した。この洗浄操作を合計３回行った後、有機層を濃縮しメチルターシャリーブチルエーテルを取り除き、１，２－ヘキサンジオール１１８．１ｇを得た。その結果、１，２－ヘキサンジオールの収率は８５．２％であり、１，２－ヘキサンジオールの純度は８５％であった。また、この１，２－ヘキサンジオールには、溶剤由来の臭気や、強アルカリ処理による着臭がみられた。

【００５４】

実験例

実施例１～９および比較例１、３で得られた１，２－アルカンジオールと、これらを精製水で２０倍に希釈した５重量％１，２－アルカンジオール水溶液を用意した。

【００５５】

１００ｍＬ容のガラス製広口瓶に、前記で用意した１，２－アルカンジオールまたは５重量％１，２－アルカンジオール水溶液を入れ、無作為に抽出した男女２０名の各パネラーに、瓶口での臭気を確認してもらい、１，２－アルカンジオールおよび５重量％１，２－アルカンジオール水溶液のそれぞれについて、以下の評価基準に基づいて評価してもらった。結果を表２に示す。なお、実施例および比較例で得られた１，２－アルカンジオール自体は、無臭の化合物である。

【００５６】

〔評価基準〕
○：ほぼ無臭である。
△：わずかに臭う。
×：臭気が明らかに感じられる。
【００５７】
次に、上記の結果に基づいて、各評価を行った者の人数を集計し、以下の基準にしたがってランク付けを行なった。なお、Ｓ以上のランクが合格である。
【００５８】
〔ランク〕
ＳＳＳ：○の評価をした評価者が１８名以上である場合
ＳＳ：○の評価をした評価者が１５～１７名であり、×の評価をした評価者がひとりもいない場合
Ｓ：○の評価をした評価者が１５～１７名であり、×の評価をした評価者がいる場合
Ａ：○の評価をした評価者が１０～１４名である場合
Ｂ：○の評価をした評価者が５～９名である場合
Ｃ：○の評価をした評価者が４名以下である場合
【００５９】
【表２】

実施例番号	臭気の有無	
	1,2-アルカンジオール	5重量%1,2-アルカンジオール水溶液
1	ＳＳ	Ｓ
2	ＳＳＳ	ＳＳ
3	ＳＳ	Ｓ
4	ＳＳ	Ｓ
5	Ｓ	Ａ
6	Ｓ	Ａ
7	ＳＳ	Ｓ
8	ＳＳ	Ｓ
9	ＳＳ	Ｓ
比較例1	Ａ	Ｂ
3	Ｂ	Ｃ

【００６０】
表１に示された結果より、実施例１～９の方法によれば、生成した１，２－アルカンジオールの臭気が比較的生じがたい従来の比較例１、３と対比しても、著しく１，２－ヘキサンジオールの臭気を低減させることができることがわかる。
【００６１】
また、実施例１～９によれば、毒性の高いベンゼンなどの溶媒を必要しないので、作業

7/31/2019, EAST Version: 4.1.2.6

環境における安全性に優れており、しかも従来、必要とされていた過酸化物のチオ硫酸ナトリウムやマンガン触媒をはじめとした過酸処理や、エステルの強アルカリによる処理などの操作を必要とせず、また酸化反応を従来よりも低い温度で行うことができるため、臭気が低減された１，２－アルカンジオールを容易にかつ効率よく安全に製造することができることがわかる。

【００６２】

したがって、本発明の製造法によれば、潤滑油、化粧品、香料、医薬品などの原料として有用な１，２－アルカンジオールを高純度で容易かつ安全に製造することができることがわかる。

【産業上の利用可能性】

【００６３】

本発明の製造法によって得られた１，２－アルカンジオールは、潤滑油、化粧品、香料、医薬品などの原料として好適に使用することができる。

7/31/2019, EAST Version: 4.1.2.6

Bibliographic data: JP2008007463 (A) — 2008-01-17

METHOD FOR PRODUCING 1,2-ALKANEDIOL

Page bookmark	JP2008007463 (A) - METHOD FOR PRODUCING 1,2-ALKANEDIOL
Inventor(s):	YAMAMOTO TATSURO; YOSHIMARU TOSHIHIKO +
Applicant(s):	OSAKA ORGANIC CHEM IND +
Classification:	- international: *C07C29/128; C07C31/20; C07B61/00*
	- cooperative:
Application number:	JP20060179791 20060629 Global Dossier
Priority number(s):	JP20060179791 20060629

Abstract of JP2008007463 (A)

PROBLEM TO BE SOLVED: To provide a method by which a low odor 1,2-alkanediol can readily and safely be produced in high purity. ;SOLUTION: The method for producing the 1,2-alkanediol is characterized as follows. An olefin is reacted with hydrogen peroxide in the presence of formic acid and the resultant reaction mixture is reacted with an alcohol. ;COPYRIGHT: (C)2008,JPO&INPIT

DESCRIPTION JP2008007463

An object of the present invention is to provide a method for easily and safely producing low odor 1,2-alkanediol with high purity. A method for producing 1,2-alkanediol, which comprises reacting an olefin with hydrogen peroxide in the presence of formic acid, and reacting the obtained reaction mixture with an alcohol. 【Selection chart】 None

Process for producing 1,2-alkanediol

[0001]

The present invention relates to a process for the preparation of 1,2-alkanediols. More particularly, it relates to a process for producing 1,2-alkanediols useful as raw materials for lubricating oils, cosmetics, perfumes, pharmaceuticals and the like.

[0002]

As a method for producing 1,2-alkanediol, a method is known in which an olefin is reacted with hydrogen peroxide in the presence of formic acid or acetic acid to

hydrolyze an epoxide produced (for example, Non-Patent Document 1 and 2). However, this method is disadvantageous in terms of production efficiency because it requires multiple steps of operation such as removal of the solvent, reaction with aqueous sodium hydroxide solution, and neutralization with hydrochloric acid after completion of the reaction.

[0003]

Further, as a method for producing 1,2-diol, there is known a method for producing 1,2-diol from an olefin via an epoxide, using peracetic acid as an oxidizing agent (see, for example, Patent Document 1). However, this method has the disadvantage that by-products of epoxide and diol are generated.

[0004]

On the other hand, as a method for producing 1,2-diol which can avoid the formation of by-products of epoxide and diol, hydrogen peroxide water is reacted with olefin in the presence of tungsten complex as a catalyst to produce 1,2-diol. Methods for producing diols are known (see, for example, non-patent documents 3 to 5). However, this method has the disadvantage that by-produced ketone and carboxylic acid, since the generated diol is further oxidized.

[0005]

There is also proposed a method of enhancing the selectivity of 1,2-diol by devising reaction conditions (see, for example, Patent Document 2 and Non-Patent Document 6). However, this method has a disadvantage in safety in the working environment because it uses highly toxic benzene as a solvent, and it is further extracted with a 30% aqueous sulfuric acid solution to neutralize the product and neutralized with a base There is a disadvantage in terms of manufacturing efficiency because it requires complicated operations.

[0006]

Also, as another method for producing 1,2-diol, a method is proposed in which an olefin and hydrogen peroxide water are reacted using a solid catalyst that can be easily separated by filtration (for example, non-patent literature 7-9). However, although this method has an improvement in the reaction operation, it has a disadvantage that the yield and selectivity of 1,2-diol are low because a large amount of epoxide and ketone are generated together with the target compound.

[0007]

Another method for producing 1,2-diols is a method for producing 1,2-diols, which comprises reacting olefins with hydrogen peroxide in the presence of a polymer compound having a sulfonic acid group. Has been proposed (see, for example, Patent Document 3). However, this method has disadvantages in terms of efficiency and safety, including reaction time.

[0008]

In addition, many conventional 1,2-alkanediols have a strong odor due to the inclusion of a trace amount of impurities, etc. Therefore, as 1,2-alkanediols used as raw materials for applications such as perfumes and cosmetics, etc. Is required to have a low odor.

[0009]

Patent Document 1: JP-A-4-41449 Patent Document 2: European Patent No. 146374 Patent Document 2: JP-A-2003-212804 Organic Syntheses, Coll.
3, 217-219 (1955) Journal of the American Chemical Society (J. Am. Chem. Soc. 67, pp. 1786-1788 (1945) Journal of Chemical Society (J. Chem. Soc. 2988-3000 (1949) Journal of American Oil Chemical Society (J. Am. Oil Chem. Soc. 44, pp. 316-320 (1967) Tetrahedron Lett. 29, pp. 823-826 (1988) Synthesis, pp. 295-297 (1989) Journal of the Catalysts (J. Cat.) 145, pp. 151-158 (1994) Chemical Communications (Chem. Commun. Pp. 325-326 (1998) J. Cat. 189, 40-51 (2000)

[0010]

The present invention has been made in view of the above-mentioned prior art, and an object of the present invention is to provide a method capable of easily and safely producing low odor 1,2-alkanediol with high purity.

[0011]

As a result of extensive research in view of the prior art, the present inventors reacted olefin with hydrogen peroxide in the presence of formic acid, and reacted the resulting reaction mixture with alcohol. It has been found that the 1,2-alkanediol is easily obtained in high purity, and the obtained 1,2-alkanediol exerts a surprising effect that hardly any odor is generated.

[0012]

The present invention has been completed based on such findings, and is characterized in that an olefin and hydrogen peroxide are reacted in the presence of formic acid, and the resulting reaction mixture is reacted with an alcohol. The present invention relates to a process for producing 2-alkanediol.

[0013]

According to the production method of the present invention, an effect is exhibited that low odor 1,2-alkanediol can be easily and safely produced with high purity.

[0014]

In the present invention, first, olefin and hydrogen peroxide are reacted in the presence of formic acid.

[0015]

As the olefin, various ones can be used in a wide range.
As a typical olefin, for example, formula (I): R <1> R <2> C = CR <3> R <4> (I) (wherein R <1>, R <2>, R <3> and R <4> each independently represent a hydrogen atom, an alkyl group which may have a substituent or an aryl group which may have a

substituent, and R <1>, And R & lt; 2 & gt ;, R & lt; 3 & gt ;, and R & lt; 4 & gt; together may form a ring). Two or more types can be mixed and used.

[0016]

As an alkyl group, a C1-C35 alkyl group is preferable and a C1-C21 alkyl group is more preferable.

As a substituent which the alkyl group may have, a hydroxyl group, a C1-C22 alkoxy group, etc. are mentioned, for example.

[0017]

As an aryl group, a C6-C35 aryl group is preferable and a C6-C22 aryl group is more preferable.

As a substituent which the aryl group may have, a hydroxyl group, a phenyl group, a C1-C22 alkoxy group etc. are mentioned, for example.

[0018]

In addition, two groups of R <1>, R <2>, R <3> and R <4> may be combined to form a ring.

Examples of such rings include cycloalkyl groups having 3 to 35 carbon atoms, and these rings may form multiple rings such as fused rings and non-fused rings with other rings.

Furthermore, a residue obtained by removing a hydrogen atom from any two of R <1>, R <2>, R <3> and R <4> is a divalent atom and / or a divalent functional group. They may be combined with each other to form a ring together with the carbon atom to which they are attached, and in this case, as a divalent atom, an oxygen atom, a nitrogen atom, a sulfur atom or the like, or Examples of the divalent functional group include a silylene group, an ethylenedioxy group, an arylenedioxy group, a carbonyl group, a sulfoxide group and a sulfone group.

[0019]

Specific examples of the olefin represented by the formula (I) include 1-hexene, 2,3-dimethyl-2-butene, 2-methyl-2-pentene, 2-heptene, cyclopentene, cyclohexene, 1 - Methylcyclohexene, cyclooctene, cyclododecene, 1-undecene, 1-dodecene, 2-cyclohexen-1-ol, 1-ethyl-1-cyclopentene, cinnamic alcohol, 2-methyl-3-phenyl-2-propene- 1-ol, 4- (1-propenyl) -1,2-dimethoxybenzene and the like can be mentioned.
[0020]

Hydrogen peroxide is usually used as an aqueous solution.
The concentration of hydrogen peroxide in the aqueous hydrogen peroxide solution is not particularly limited, but is preferably 1 to 80%, and more preferably 3 to 60%.
[0021]

The amount of hydrogen peroxide is preferably equal to or less than the equivalent to the olefin double bond. Usually, when the amount of hydrogen peroxide is excessive to the olefin, it is necessary to treat the excess peroxide after the reaction, and in some cases, it is removed by washing or filtration of the treatment. A process is also required, and an odorant is generated or a side reaction product is generated by a peroxide treating agent (usually, in the case of sodium thiosulfate, etc., an odor from the sulfur source is easily generated) In the case of catalysts such as manganese oxide, there are many side reactions such as oxidation of diol, and there are many cases where odor components and impurities are easily generated due to heat generation, and safety aspects due to increased risk due to excess peroxide. Therefore, it is preferably 0.5 to 1 equivalent, more preferably 0.7 to 0.95 equivalent to the olefin double bond.
[0022]

The amount of formic acid safely causes the oxidation of the olefin under milder conditions, so that the formic acid is generated in the reaction system, and the side reaction between the epoxy compound formed as an intermediate from the olefin and the diol is suppressed. Therefore, from the viewpoint of reacting an epoxy intermediate with formic acid to open the ring, it is usually 1 to 30 equivalents, more preferably 1 to

15 equivalents, more preferably from the viewpoint of production efficiency to the olefin double bond. Preferably, it is 1 to 5 equivalents.

[0023]

The reaction of an olefin and hydrogen peroxide can be easily carried out, for example, by mixing the olefin and formic acid, and then adding hydrogen peroxide to the mixture while stirring the obtained mixture.

[0024]

The reaction temperature of the olefin and hydrogen peroxide is usually 20 to 100 ° C, preferably 30 to 60 ° C.

[0025]

A solvent may be used in the reaction of the olefin and hydrogen peroxide.
Examples of the solvent include ethers, esters, nitriles, amides, sulfoxides, hydrocarbon compounds and the like.
The amount of the solvent is not particularly limited, but generally, it is preferably 1000 parts by volume or less, more preferably 300 parts by volume or less, per 100 parts by volume of the olefin.

[0026]

The end point of the reaction between the olefin and the hydrogen peroxide can be easily confirmed by, for example, analysis by gas chromatography and confirmation of disappearance of the peracid by titration of the peroxide.

[0027]

Thus, the reaction of the olefin with hydrogen peroxide gives a reaction mixture containing the ester formed.
The resulting reaction mixture may be concentrated if necessary.

[0028]

Next, the reaction mixture obtained is reacted with an alcohol.
Such a reaction is specifically a transesterification reaction of the formed ester contained in the obtained reaction mixture with an alcohol.
[0029]

As alcohol, a C1-C4 linear or branched monohydric alcohol is mentioned, for example. Specific examples of such monohydric alcohols include methanol, ethanol, isopropanol and the like, but the present invention is not limited to such examples.
[0030]

The amount of alcohol is the total of the ester portion of the formed ester contained in the reaction mixture and the remaining formic acid, from the viewpoint of completely formicifying the ester site contained in the reaction mixture and the remaining formic acid. Preferably, it is 1 to 10 equivalents, more preferably 2 to 10 equivalents, still more preferably 3 to 5 equivalents.
[0031]

In addition, a catalyst can be used in the case of reaction with the obtained reaction mixture and alcohol.
As a catalyst, what is generally used in the case of transesterification may be used. Examples of the catalyst include sulfonyl compounds such as sulfuric acid and paratoluenesulfonic acid, acid catalysts such as polymer compounds having a sulfonic acid group, metals such as lithium compounds, titanium compounds, sodium compounds, aluminum compounds, tin compounds, zinc compounds and the like Although a catalyst etc. are mentioned, this invention is not limited only to this illustration.
[0032]

The amount of the catalyst is not particularly limited, but generally it is preferably 0.01 to 5 parts by weight, more preferably 0.05 to 1 part by weight, per 100 parts by weight of the reaction mixture.

[0033]

Moreover, although reaction of an olefin and hydrogen peroxide can also be normally performed without using a solvent, it is also possible to use a solvent.
As the solvent, those generally used in oxidation reaction and transesterification reaction can be used. Examples of the solvent include ethers, nitriles, amides, sulfoxides, hydrocarbon compounds and the like. The amount of the solvent is not particularly limited, but generally, it is preferably 50 to 1000 parts by volume, more preferably 100 to 300 parts by volume, per 100 parts by volume of the olefin.
[0034]

The reaction of the obtained reaction mixture with the alcohol can be carried out, for example, by adding and mixing an alcohol and, if necessary, a catalyst to the reaction mixture.
[0035]

The reaction temperature is preferably in the range of usually 20 to 80 ° C., and from the viewpoint of advancing the equilibrium reaction by selectively taking out the formic acid ester to be produced out of the reaction system, the boiling point or less of the alcohol used and it is more preferable that it is more than the boiling point of the formic acid ester to be produced.
During the reaction, it is preferable to recover the formic acid ester to be produced, but in particular, when methanol is used as the alcohol, methyl formate having a boiling point lower than that of other formate esters is generated, so that it is milder. The reaction of the reaction mixture with the alcohol can be carried out under the conditions, and since methanol and methyl formate do not azeotrope and only methyl formate volatilizes, methyl formate can be easily removed out of the reaction system.
[0036]

The reaction of the reaction mixture with the alcohol can usually be carried out without using a solvent, but it is also possible to use a solvent. As the solvent, those used in

transesterification can be used. Examples of the solvent include ethers, nitriles, amides, sulfoxides, hydrocarbon compounds and the like.

[0037]

The end point of the reaction of the reaction mixture with the alcohol can be easily confirmed, for example, by analysis by gas chromatography.

[0038]

Thus, the target compound 1,2-alkanediol is obtained.
When a catalyst is used in the reaction, it is preferable to neutralize the catalyst by adding an alkali to the reaction mixture containing the formed 1,2-alkanediol.

[0039]

The formate ester, formic acid and alcohol contained in the reaction mixture containing 1,2-alkanediol can be recovered by concentrating the reaction mixture and reused. The obtained 1,2-alkanediol may be purified by, for example, distillation. In addition, when the 1,2-alkanediol exhibits such a water solubility that allows at least a transition to the aqueous layer when it is mixed with water, the 1,2-alkanediol is selected from normal hexane, cyclohexane, 2- By mixing with a hydrophobic solvent such as methyl hexane and water, the odor component is transferred to the organic layer, and the 1,2-alkanediol is transferred to the aqueous layer, thereby reducing the odor and reducing the 1,2-alkanediol to water It can be extracted into layers.

[0040]

The production method of the present invention is excellent in safety in the working environment because it does not require highly toxic solvents such as benzene. Moreover, according to the production method of the present invention, 1,2-alkanediol can be easily obtained in high purity and high yield. Furthermore, since the 1,2-alkanediol obtained by the production method of the present invention hardly emits an odor, it can be suitably used as a raw material for lubricating oils, cosmetics, perfumes, pharmaceuticals and the like.

[0041]

Next, the present invention will be described in more detail based on examples, but the present invention is not limited to such examples.

[0042]

Example 1 (1) Reaction of Olefin with Hydrogen Peroxide 100 g of 1-hexene and 237.5 g of formic acid (purity 95%) are mixed in a 1 liter beaker, and the resulting mixture is heated with stirring. When reaching 30 ° C., 98.2 g of a 35% aqueous hydrogen peroxide solution was slowly added to the mixture over 10 hours.
Since the reaction exothermed, the mixture was externally cooled to a temperature of 30 to 40 ° C.

[0043]

After completion of the addition of the aqueous hydrogen peroxide solution, the reaction was terminated by maintaining the temperature of the mixture at 35 ° C. for 2 hours. The end point of the reaction was confirmed by titration of the amount of peroxide (potassium iodide titration method). The resulting reaction mixture is concentrated until the amount of excess formic acid is distilled off (there is no need to completely concentrate the formic acid because it is also a formic acid ester), and the concentrated reaction mixture is subjected to the next alcohol Used for reaction.

[0044]

(2) Reaction of reaction mixture and alcohol 176.7 g of the concentrated reaction mixture obtained in the above (1), 170 g of methanol and 0.09 g of p-toluenesulfonic acid monohydrate are mixed in a 1 liter beaker The resulting methyl formate was stirred at 55 ° C. for 7 hours while being removed from the beaker. Thereafter, 0.08 g of a 30% aqueous solution of sodium hydroxide is added to the beaker to neutralize the catalyst (p-toluenesulfonic acid monohydrate), and then methanol is distilled off under reduced pressure to obtain 1,2- 120.7 g of hexanediol was obtained. As a result, the yield of 1,2-hexanediol was 92%, and the purity of 1,2-hexanediol was 92%.

[0045]

Next, when the obtained 1,2-hexanediol is purified by distillation, the yield is 76%, and by further re-distilling the initial distillation, the purity is 98% or more with a distillation yield of 94% 1, 2-Hexanediol was obtained.

[0046]

Example 2 1, 2-Hexanediol was prepared as in Example 1 and was purified to distillation.

5 g (purity 98%) of the obtained 1,2-hexanediol, 10 g of normal hexane and 10 g of water were mixed and stirred at 25 ° C. Separate the hexane layer and concentrate the aqueous layer to remove the odor component, 4.9 g of 1,2-hexanediol (98% yield, relative to 1,2-hexanediol mixed with normal hexane and water, purity 99 %) Got.

[0047]

Examples 3 to 9 In the same manner as in Example 1, except that olefins shown in Table 1 were used instead of 1-hexene and formic acid, hydrogen peroxide aqueous solution etc. were used in the amounts shown in Table 1, An alkanediol is obtained. The obtained 1,2-alkanediol and its yield and purity are shown in Table 1.

[0048]

[0049]

Comparative Example 1 41 g of cyclohexene is charged into a glass flask and heated to 30 ° C. while stirring.

When 30 ° C. was reached, 30% peracetic acid was slowly added over a period of 2 hours to a molar ratio of 1.2 times. Since the reaction is exothermic, the mixture was cooled from the outside so that the temperature of the mixture became about 30 ° C. After completion of the peracetic acid addition, the reaction was completed by maintaining the temperature of the mixture at 30 ° C. for 1 hour. The reaction crude

solution was analyzed by gas chromatography to confirm that cyclohexene oxide was obtained in 93% of the yield.

[0050]

50 g of water was charged into a glass flask and heated with stirring, and when reaching 70 ° C., 21.6 g of the solution containing cyclohexene oxide obtained above and acetic acid was slowly added over 2 hours. After completion of the addition of the solution, the reaction crude solution was immediately analyzed by gas chromatography. As a result, cyclohexane-1,2-diol was obtained in 94% yield based on cyclohexene oxide and 87.4% based on cyclohexene It confirmed that it was obtained by the yield.

[0051]

Comparative Example 2 500 mg of Nafion-NR50 (manufactured by DuPont) (equivalent weight of sulfonic acid group to olefin is equivalent to 0.04), 2.23 g (19.6 mmol) of a 30% aqueous hydrogen peroxide solution and 825 mg of 1-hexene (9. 81 mmol) were mixed and stirred at 70 ° C. for 24 hours. As a result of analyzing the reaction solution by gas chromatography, almost no progress of the reaction was observed.

[0052]

The reaction solution was filtered to remove Nafion-NR50, and 10 mg (0.115 mmol) of manganese dioxide was added to decompose excess hydrogen peroxide present in the solution. The reaction solution was filtered and then water and the like were distilled off using a rotary evaporator to obtain 1,2-hexanediol as a concentrated mixture (yield: less than 5%).

[0053]

Comparative Example 3 In 176.7 g of the concentrated reaction mixture obtained in the same manner as the reaction of (1) olefin and hydrogen peroxide in Example 1, 239.3 g of a 30% aqueous solution of sodium hydroxide was added to 20 to 35 It dripped at ° C. The end point of the reaction was confirmed by analysis by gas chromatography. To the obtained reaction mixture, 142.4 g of methyl tertiary butyl ether as an extraction solvent

was added, and after stirring, the mixture was allowed to stand to separate an aqueous layer containing potassium formate. To the organic layer, 71.2 g of 20% brine was added and washed, and after stirring and standing, the aqueous layer was separated. After the washing operation was performed a total of three times, the organic layer was concentrated to remove methyl tertiary butyl ether to obtain 118.1 g of 1,2-hexanediol. As a result, the yield of 1,2-hexanediol was 85.2%, and the purity of 1,2-hexanediol was 85%. Moreover, the odor derived from a solvent and the odor by strong alkali treatment were seen by this 1, 2- hexanediol.

[0054]

Experimental Example The 1,2-alkanediols obtained in Examples 1 to 9 and Comparative Examples 1 and 3 and a 5 wt% 1,2-alkanediol aqueous solution obtained by diluting them with purified water 20 times were prepared.

[0055]

In a 100 mL glass jar, put the above-prepared 1,2-alkanediol or 5% by weight 1,2-alkanediol aqueous solution, and randomly extracted each panel of 20 men and women by using a bottle neck. The odor was confirmed, and each of the 1,2-alkanediol and the 5 wt% 1,2-alkanediol aqueous solution was evaluated based on the following evaluation criteria.

The results are shown in Table 2. The 1,2-alkanediol itself obtained in Examples and Comparative Examples is an odorless compound.

[0056]

[Evaluation Criteria] ○: Almost odorless. :: Slightly smelly. X: The odor is clearly felt.

[0057]

Next, based on the above results, the number of persons who performed each evaluation was counted, and ranking was performed according to the following criteria. In addition, the rank of S or more is a pass.

[0058]

[Rank] SSS: When there are 18 or more evaluators who have evaluated ○ SS: There are 15 to 17 evaluators who have evaluated ○, and there is no one who has evaluated × S: When there are 15 to 17 evaluators who have a rating of ○ and there are evaluators who have a rating of A A: When there are 10 to 14 evaluators who have a rating of B B: Evaluation that has a rating of ○ When there are 5 to 9 persons C: If there are 4 or less evaluators who have made an evaluation

[0059]

[0060]

From the results shown in Table 1, according to the methods of Examples 1 to 9, the odor of the produced 1,2-alkanediol is relatively unlikely to be generated, even in comparison with the conventional Comparative Examples 1 and 3. It can be seen that the odor of 1,2-hexanediol can be reduced.

[0061]

Further, according to Examples 1 to 9, since a solvent such as highly toxic benzene is not required, the safety in the working environment is excellent, and the sodium thiosulfate and manganese of the peroxide conventionally required are required. There is no need for operations such as treatment with peracid including catalyst or treatment with strong alkali of ester, and the oxidation reaction can be performed at a temperature lower than conventional, so the odor is reduced 1,2-alkane It can be seen that diols can be produced easily and efficiently safely.

[0062]

Therefore, it is understood that according to the production method of the present invention, 1,2-alkanediol useful as a raw material for lubricating oil, cosmetics, perfumes, pharmaceuticals and the like can be easily and safely produced with high purity.

[0063]

The 1,2-alkanediol obtained by the production method of the present invention can be suitably used as a raw material for lubricating oils, cosmetics, perfumes, pharmaceuticals and the like.

CLAIMS JP2008007463

1.

1. A process for producing 1,2-alkanediol, which comprises reacting an olefin with hydrogen peroxide in the presence of formic acid, and reacting the resulting reaction mixture with an alcohol.

2.

The olefin is represented by the formula: R <1> R <2> C = CR <3> R <4> (I) (wherein R <1>, R <2>, R <3> and R <4> are And each independently represents a hydrogen atom, an alkyl group which may have a substituent, or an aryl group which may have a substituent, and R <1>, R <2>, R <3> And R <4> may form a ring together) and an olefin represented by the formula:

3.

The method according to claim 1 or 2, wherein the amount of hydrogen peroxide is 0.5 to 1 equivalent based on the olefin double bond.

PATENT COOPERATION TREATY

From the INTERNATIONAL SEARCHING AUTHORITY

To: Lynda L. Calderone
Calderone Bullock LLC
One Greentree Centre
9000 Lincoln Drive East, Suite 301
Marlton, New Jersey 08053
United States of America

PCT

NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL SEARCH REPORT AND THE WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY, OR THE DECLARATION

(PCT Rule 44.1)

	Date of mailing *(day/month/year)* 19 APR 2019
Applicant's or agent's file reference I0113-198WO	**FOR FURTHER ACTION** See paragraphs 1 and 4 below
International application No. PCT/US19/15928	International filing date *(day/month/year)* 30 January 2019 (30.01.2019)
Applicant INOLEX INVESTMENT CORPORATION	

1. ☒ The applicant is hereby notified that the international search report and the written opinion of the International Searching Authority have been established and are transmitted herewith.

   Filing of amendments and statement under Article 19:
   The applicant is entitled, if he so wishes, to amend the claims of the international application (see Rule 46):

   When? The time limit for filing such amendments is normally two months from the date of transmittal of the international search report.

   How? Directly to the International Bureau of WIPO preferably through ePCT or on paper to, 34 chemin des Colombettes 1211 Geneva 20, Switzerland, Facsimile No.: +41 22 338 82 70

   For more detailed instructions, see *PCT Applicant's Guide*, International Phase, paragraphs 9.004 – 9.011.

2. ☐ The applicant is hereby notified that no international search report will be established and that the declaration under Article 17(2)(a) to that effect and the written opinion of the International Searching Authority are transmitted herewith.

3. ☐ **With regard to any protest** against payment of (an) additional fee(s) under Rule 40.2, the applicant is notified that:
   ☐ the protest together with the decision thereon has been transmitted to the International Bureau together with any request to forward the texts of both the protest and the decision thereon to the designated Offices.
   ☐ no decision has been made yet on the protest; the applicant will be notified as soon as a decision is made.

4. **Reminders**

   The applicant may **submit comments on an informal basis on the written opinion of the International Searching Authority** to the International Bureau. These comments will be made available to the public after international publication. The International Bureau will send a copy of such comments to all designated Offices unless an international preliminary examination report has been or is to be established.

   Shortly after the expiration of **18 months from the priority date, the international application will be published** by the International Bureau. If the applicant wishes to avoid or postpone publication, a notice of withdrawal of the international application, or of the priority claim, must reach the International Bureau before the completion of the technical preparations for international publication (Rules 90*bis*.1 and 90*bis*.3).

   Within **19 months** from the priority date, but only in respect of some designated Offices, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase **until 30 months** from the priority date (in some Offices even later); otherwise, the applicant must, **within 20 months** from the priority date, perform the prescribed acts for **entry into the national phase** before those designated Offices. In respect of other designated Offices, the time limit of **30 months** (or later) will apply even if no demand is filed within 19 months. For details about the applicable time limits, Office by Office, see www.wipo.int/pct/en/texts/time_limits.html and the *PCT Applicant's Guide*, National Chapters.

   Within **22 months from the priority date, the applicant may request that a supplementary international search be carried out** by a different International Searching Authority that offers this service (Rule 45*bis*.1). The procedure for requesting supplementary international search is described in the *PCT Applicant's Guide*, International Phase, paragraphs 8.006-8.032.

Name and mailing address of the ISA/US Mail Stop PCT, Attn: ISA/US Commissioner for Patents P.O. Box 1450, Alexandria, Virginia 22313-1450 Facsimile No. 571-273-8300	Authorized officer Shane Thomas Telephone No. PCT Helpdesk: 571-272-4300 PCT OSP: 571-272-7774

Form PCT/ISA/220 (July 2017)

PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference I0113-198WO	FOR FURTHER ACTION	see Form PCT/ISA/220 as well as, where applicable, item 5 below.
International application No. PCT/US19/15928	International filing date *(day/month/year)* 30 January 2019 (30.01.2019)	(Earliest) Priority Date *(day/month/year)* 30 January 2018 (30.01.2018)
Applicant INOLEX INVESTMENT CORPORATION		

This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 9 sheets.

☐ It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**

a. With regard to the **language**, the international search was carried out on the basis of:

☒ the international application in the language in which it was filed.

☐ a translation of the international application into _____ which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b)).

b. ☐ This international search report has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43.6*bis*(a)).

c. ☐ With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, see Box No. I.

2. ☐ **Certain claims were found unsearchable** (see Box No. II).

3. ☐ **Unity of invention is lacking** (see Box No. III).

4. With regard to the **title**,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

☒ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 38.2, by this Authority as it appears in Box No. IV. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. With regard to the **drawings**,

a. the figure of the **drawings** to be published with the abstract is Figure No. 1

☒ as suggested by the applicant.

☐ as selected by this Authority, because the applicant failed to suggest a figure.

☐ as selected by this Authority, because this figure better characterizes the invention.

b. ☐ none of the figures is to be published with the abstract.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US19/15928

A. CLASSIFICATION OF SUBJECT MATTER

IPC - A61K 8/34; A01N 31/00, 31/02; C07C 5/00, 11/10, 11/107; C10G 3/00; B01J 21/00 (2019.01)
CPC - B01J 21/00, 21/005, 21/02, 21/04, 23/00, 23/005, 23/06; C07C 5/00, 11/10, 11/107; C10G 3/00, 3/42, 3/44, 3/45; A61K 8/34, 8/345; A01N 31/00, 31/02

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
See Search History document

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
See Search History document

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
See Search History document

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X --- A	WO 2016/007196 A1 (COBALT TECHNOLOGIES, INC.) 14 January 2016; paragraphs [22]-[23], [33]-[34], [51], [54], [90], [0103], [122]; claims 1, 29-30, 48; figures 11, 13	35-41 --- 1, 30, 42, 56, 65, 72, 81
A	US 2006/0161025 A1 (OSTERHOLT, C et al.) 20 July 2006; paragraphs [0085]-[0096]	1, 30, 42, 56, 65, 72, 81
A	US 2016/0090331 A1 (IFP ENERGIES NOUVELLES) 31 March 2016; paragraphs [0025]-[0029], [0132]-[0136]; claim 1	1, 30, 42, 56, 65, 72, 81
A	DAI, L et al. Directing enzyme devolution for biosynthesis of alkanols and 1, n-alkanediols from natural polyhydroxy compounds. Metabolic engineering. 1 November 2017. Vol. 44; page 77, figure 7a	1, 30, 42, 56, 65, 72, 81

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" earlier application or patent but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
18 March 2019 (18.03.2019)	19 APR 2019
Name and mailing address of the ISA/ Mail Stop PCT, Attn: ISA/US, Commissioner for Patents P.O. Box 1450, Alexandria, Virginia 22313-1450 Facsimile No. 571-273-8300	Authorized officer Shane Thomas PCT Helpdesk: 571-272-4300 PCT OSP: 571-272-7774

Form PCT/ISA/210 (second sheet) (January 2015)

PATENT COOPERATION TREATY

From the
INTERNATIONAL SEARCHING AUTHORITY

To: Lynda L. Calderone
Calderone Bullock LLC
One Greentree Centre
9000 Lincoln Drive East, Suite 301
Marlton, New Jersey 08053
United States of America

PCT

WRITTEN OPINION OF THE
INTERNATIONAL SEARCHING AUTHORITY

(PCT Rule 43*bis*.1)

Date of mailing *(day/month/year)* 19 APR 2019

Applicant's or agent's file reference
I0113-198WO

FOR FURTHER ACTION
See paragraph 2 below

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/US19/15928	30 January 2019 (30.01.2019)	30 January 2018 (30.01.2018)

International Patent Classification (IPC) or both national classification and IPC
IPC - A61K 8/34; A01N 31/00, 31/02; C07C 5/00, 11/10, 11/107; C10G 3/00; B01J 21/00 (2019.01)
CPC - B01J 21/00, 21/005, 21/02, 21/04, 23/00, 23/005, 23/06; C07C 5/00, 11/10, 11/107; C10G 3/00, 3/42, 3/44, 3/45; A61K 8/34, 8/345; A01N 31/00, 31/02

Applicant INOLEX INVESTMENT CORPORATION

1. This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☐ Box No. II Priority
- ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Rule 43*bis*.1(a)(i) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☒ Box No. VII Certain defects in the international application
- ☒ Box No. VIII Certain observations on the international application

2. **FURTHER ACTION**

If a demand for international preliminary examination is made, this opinion will be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA") except that this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1*bis*(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of 3 months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

Name and mailing address of the ISA/US	Date of completion of this opinion	Authorized officer
Mail Stop PCT, Attn: ISA/US Commissioner for Patents P.O. Box 1450, Alexandria, Virginia 22313-1450 Facsimile No. 571-273-8300	18 March 2019 (18.03.2019)	Shane Thomas PCT Helpdesk: 571-272-4300 PCT OSP: 571-272-7774

Form PCT/ISA/237 (cover sheet) (January 2015)

WRITTEN OPINION OF THE
INTERNATIONAL SEARCHING AUTHORITY

International application No.
PCT/US18/15928

Box No. I Basis of this opinion

1. With regard to the **language**, this opinion has been established on the basis of:

   ☒ the international application in the language in which it was filed.

   ☐ a translation of the international application into _____ which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b)).

2. ☐ This opinion has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43*bis*.1(a)).

3. ☐ With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, this opinion has been established on the basis of a sequence listing:

   a. ☐ forming part of the international application as filed:

      ☐ in the form of an Annex C/ST.25 text file.

      ☐ on paper or in the form of an image file.

   b. ☐ furnished together with the international application under PCT Rule 13*ter*.1(a) for the purposes of international search only in the form of an Annex C/ST.25 text file.

   c. ☐ furnished subsequent to the international filing date for the purposes of international search only:

      ☐ in the form of an Annex C/ST.25 text file (Rule 13*ter*.1(a)).

      ☐ on paper or in the form of an image file (Rule 13*ter*.1(b) and Administrative Instructions, Section 713).

4. ☐ In addition, in the case that more than one version or copy of a sequence listing has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that forming part of the application as filed or does not go beyond the application as filed, as appropriate, were furnished.

5. Additional comments:

WRITTEN OPINION OF THE
INTERNATIONAL SEARCHING AUTHORITY

International application No.
PCT/US19/15928

Box No. V **Reasoned statement under Rule 43*bis*.1(a)(i) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement**

1. Statement

Novelty (N)	Claims	1-34, 42-87	YES
	Claims	35-41	NO
Inventive step (IS)	Claims	1-34, 42-87	YES
	Claims	35-41	NO
Industrial applicability (IA)	Claims	1-87	YES
	Claims	NONE	NO

2. Citations and explanations:

Claims 35-41 lack novelty under PCT Article 33(2) as being anticipated by WO 2016/007196 A1 to COBALT TECHNOLOGIES, INC. (hereinafter 'Cobalt').

Regarding claim 35, Cobalt discloses a method for treating a catalyst for use in dehydration of alcohols (a method for dehydrating an alcohol to a diene using a catalyst; paragraph [40]; claim 1), comprising providing a gamma-alumina catalyst; treating the gamma-alumina catalyst with a base comprising a Group I or a Group II metal (providing gamma-alumina catalyst treated with Group 1 or Group 2 metal promoters (base); paragraphs [33]-[34], [122]; claims 1, 29); and heating the gamma-alumina catalyst to a temperature of about 400 degree C to about 500 degree C (calcining the treated gamma-alumina catalyst at 100-900 degrees C; paragraphs [33]-[34], [122]).

Regarding claim 36, Cobalt discloses the method according to claim 35, and further discloses wherein the base comprises calcium (treating gamma-alumina catalyst with calcium metal promoters (base); paragraphs [33]-[34], [122]; claim 30).

Regarding claim 37, Cobalt discloses the method according to claim 36, and further discloses wherein the base is a calcium promoter and the heating occurs during calcination (treating gamma-alumina catalyst with calcium metal promoter (base) followed by calcination; paragraphs [33]-[34], [122]).

Regarding claim 38, Cobalt discloses the method according to claim 37, and further discloses wherein the calcium promoter includes is used in an amount of about 0.01 weight percent to about 4 weight percent based on the weight of CaO determined after calcination (Ca-modified catalyst comprising 0.8-4.0 wt. % of Ca+2 deposited on the catalyst support due to calcination (consistent with 1.1-5.6 weight percent based on the weight of CaO, provided the molar mass of Ca is 40g/mol and CaO is 56 g/mol); paragraph [122]).

Regarding claim 39, Cobalt discloses the method according to claim 38, and further discloses wherein the calcium promoter is used in an amount of about 1 weight percent to about 2 weight percent based on the weight of CaO determined after calcination (Ca-modified catalyst comprising 0.8-4.0 wt. % of Ca+2 deposited on the catalyst support due to calcination (consistent with 1.1-5.6 weight percent based on the weight of CaO, provided the molar mass of Ca is 40g/mol and CaO is 56 g/mol); paragraph [122]).

Regarding claim 40, Cobalt discloses the method according to claim 35, and further discloses wherein the temperature during calcination is about 420 degree C to about 480 degree C (calcining the treated gamma-alumina catalyst at 100-900 degrees C; paragraph [122]).

Regarding claim 41, Cobalt discloses the method according to claim 40, and further discloses wherein the temperature during calcination is about 440 degree C to about 460 degree C (calcining the treated gamma-alumina catalyst at 100-900 degrees C; paragraph [122]).

Claim 1 meets the criteria set out in PCT Article 33(2)-(3), because the prior art does not teach or fairly suggest the claimed matter.

Regarding claim 1, US 2006/0161025 A1 to OSTERHOLT, et al. (hereinafter 'Osterholt') discloses a process for the synthesis of 1,2-alkanediols (a process for synthesis of 1,2-hexanediol or 1,2-octanediol; paragraphs [0085]-[0096]), comprising: providing an alkene having a carbon chain of about 5 to about 20 carbon atoms; and converting the alkene to a 1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms (providing 1-hexene and converting it into 1,2-hexanediol, or providing 1-octene and converting it to 1,2-octanediol; paragraphs [0085]-[0096]).

Where Osterholt discloses chemical synthesis of 1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms by conversion of alkene (Osterholt, paragraphs [0085]-[0096]), Osterholt fails to disclose a bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms synthesized by conversion of a bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

-***-Continued Within the Next Supplemental Box-***-

WRITTEN OPINION OF THE
INTERNATIONAL SEARCHING AUTHORITY

International application No.
PCT/US19/15928

Box No. VII Certain defects in the international application

The following defects in the form or contents of the international application have been noted:

Claim 40 is objected to under PCT Rule 66.2(a)(iii) as containing the following defect in the form or contents thereof: in the line 1 of claim 40, the phrase "The process according to claim 35, temperature", as best understood, is interpreted as "The process according to claim 35, wherein the temperature" for the purpose of this opinion.

WRITTEN OPINION OF THE
INTERNATIONAL SEARCHING AUTHORITY

International application No.
PCT/US19/15928

Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

Claims 39-41 are objected to under PCT Rule 66.2(a)(v) as lacking clarity under PCT Article 6 because claims 39-41 are indefinite for the following reason(s): Claims 39-41 refer to "the process according to claim", in line 1. There is a lack of antecedent basis for these limitations in the claim. For the purpose of this opinion, as best understood, the claims are interpreted to read "the method according to claim".

Supplemental Box

In case the space in any of the preceding boxes is not sufficient.
Continuation of:

-***-Continued from Box V: Citations and Explanations-***-

Cobalt discloses a process for the synthesis of bio-alkenes (a process of biomass conversion to bio-dienes (bio-alkenes); figure 1; paragraph [94]), comprising: synthesizing a bio-alkene having a carbon chain of about 4 carbon atoms and a bio-alkene regioselectivity of at least about 80% (dehydration of bio-alcohol to bio-1,3-butadiene with 90-99% regioselectivity; paragraphs [54], [90], [103]); and converting a bio-alcohol to a bio-1,3-alkene having a carbon chain of about 4 carbon atoms (dehydration of bio-alcohol to bio-1,3-butadiene; figure 13; paragraphs [22]-[23]). Cobalt also discloses synthesizing a bio-alkene having a carbon chain of about 4 carbon atoms and a bio-1-alkene yield of at least about 80% (synthesis of 1-butene using catalyst having 80% yield; figure 11; paragraph [51]).

Where Cobalt teaches a process for production of bio-alkenes with high regioselectivity (Cobalt; paragraphs [54], [90], [103]), the process of Cobalt is drawn to the synthesis of bio-alkenes from bio-diols, rather than synthesis of bio-1,2-alkanediols from bio-alkenes. Moreover, the bio-alkene of Cobalt has a shorter chain of only 4 carbon atoms (Cobalt; figures 11, 13; paragraphs [51], [54]).

US 2016/0090331 A1 to IFP ENERGIES NOUVELLES (hereinafter 'Energies') discloses a process for the synthesis of bio-alkanediols, comprising: providing a bio-carbohydrate having a carbon chain of about 5 to about 20 carbon atoms (a process for converting biomass comprising $(C_6H_{10}O_5)_n$ carbohydrates to alkanediols (bio-alkanediols), such as pentanediols and hexanediols, from a long list of compounds; paragraphs [0025]-[0029], [0132]-[0138]; claim 1); and converting the bio-carbohydrate to a bio-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms (converting biomass to alkanediols (bio-alkanediols), such as pentanediols and hexanediols, from a long list of compounds; paragraphs [0025]-[0029], [0132]-[0138]; claim 1).

Where Energies discloses synthesis of bio-alkanediols having a carbon chain of about 5 to about 20 from a bio-carbohydrate (Energies; paragraphs [0025]-[0029], [0132]-[0138]; claim 1), Energies fails to disclose a bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms synthesized by conversion of a bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

The document entitled 'Directing enzyme devolution for biosynthesis of alkanols and 1,n-alkanediols from natural polyhydroxy compounds' to DAI, et al. (hereinafter 'Dai') discloses a process for the synthesis of bio-1,2-alkanediols (synthesis of 1,2-pentanediol from biomass (bio-1,2-alkanediol); page 77, figure 7A, steps 1-3), comprising: providing a bio-polyhydroxy compounds having a carbon chain of about 5 to about 20 carbon atoms; and converting the bio-polyhydroxy compounds to a bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms (converting polyhydroxy compounds derived from biomass to 1,2-pentanediol (bio-1,2-alkanediol); page 77, figure 7 A, step 1-3).

Where Dai discloses a process for the synthesis of bio-1,2-alkanediols, comprising: providing a bio-polyhydroxy compounds having a carbon chain of about 5 to about 20 carbon atoms; and converting the bio-polyhydroxy compounds to a bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms, Dai fails to teach a bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms synthesized by conversion of a bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

Therefore, the modification of providing for a process for the synthesis of bio-1,2-alkanediols, comprising: providing a bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%; and converting the bio-alkene to a bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms would not have been obvious to one of ordinary skill in the art at the time of the invention, provided the previous disclosures of Osterholt, Cobalt, Energies, and Dai, as none of the disclosures, taken alone or in combination, teach a 1,2-alkanediols with a carbon atom chain larger than 4, and obtained from a naturally-derived alkene having high bio-1-alkene regioselectivity.

Claims 30, 42, 56, 65, 72, and 81 also meet the criteria set out in PCT Article 33(2)-(3), because claims 30, 42, 56, 65, 72, and 81 share the core inventive element of claim 1, namely, a bio-1,2-alkanediol having a carbon chain length of about 5 to about 20 carbon atoms synthesized by conversion of a bio-alkene having a carbon chain of about 5 to about 20 carbon atoms and a bio-1-alkene regioselectivity of at least about 80%.

Claims 2-29, 31-34, 43-55, 57-64, 66-71, 73-80, and 82-87 also meet the criteria set out in PCT Article 33(2)-(3), because they depend upon claims 30, 42, 56, 65, 72, and 81, respectively.

Claims 1-87 have industrial applicability as defined by PCT Article 33(4) because the subject matter can be made or used in industry.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor: **Rocco V. Burgo**	Attorney Docket No: **INLX.0198U2/484799**
Application No.: **16/262,902**	Confirmation No.: **1028**
Filing Date: **January 20, 2019**	Art Unit: **1612**
Title: ***Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same***	Examiner: **HOLLOMAN, Nannette**

VIA EFS

November 11, 2021

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

INFORMATION DISCLOSURE STATEMENT

Pursuant to 37 C.F.R. §§ 1.56 and in accordance with 37 C.F.R. §§ 1.97 and 1.98, information relating to the above-identified application is hereby disclosed. The Examiner in charge of the above-identified application is requested to consider and make of record the references listed on PTO Forms SB/08A and SB/08B, formerly known as PTO Form 1449, submitted herewith.

Inclusion of the information submitted herewith is not to be construed as an admission that the information is material as that term is defined in 37 C.F.R. § 1.56(b).

In accordance with 37 C.F.R. § 1.97(g), the filing of this Information Disclosure Statement shall not be construed to mean that a search has been made.

This Information Disclosure Statement is being filed:

☐ within three months of the filing date of the patent application.

☐ within three months of the date of entry into the national stage as set forth in 37 C.F.R. § 1.491 of the international application.

☐ **before** the mailing date of a first Office Action on the merits.

☐ **before** the mailing date of a first Office Action on the merits after the filing of a Request for Continued Examination under § 1.114..

☒ **after** the mailing date of a first Office Action on the merits or after the filing of a Request for Continued Examination under § 1.114, but before the mailing date of a Final Office Action under 37 C.F.R. §

1.116 or a Notice of Allowance under 37 C.F.R. § 1.311, and accordingly is accompanied by:

☐ the Statement under 37 C.F.R. § 1.97(e) (see "Statement" below); or

☒ the Fee under 37 C.F.R. § 1.17(p) (see below); or

☐ No fee is owed by the applicant(s).

☐ In accordance with 37 C.F.R. § 1.129(a), this Information Disclosure Statement is being filed in connection with ☐ the first or ☐ second After Final Submission, and accordingly is accompanied by the Statement under 37 C.F.R. § 1.97(e) (see "Statement" below) and the Fee under 37 C.F.R. § 1.17(p) (see below).

☐ **after** the mailing date of a Final Office Action under 37 C.F.R. § 1.116 or a Notice of Allowance under 37 C.F.R. § 1.311, but before, or simultaneously with, the payment of the Issue Fee, and accordingly is accompanied by the Statement under 37 C.F.R. § 1.97(e) (see below) and the Fee under 37 C.F.R. § 1.17(p) (see below).

☒ Copies of the foreign patent documents and non-patent literature references listed on the attached PTO Forms SB/08a and SB/08b, formerly known as PTO Form 1449, are enclosed herewith.

EXCEPT THAT:

☐ In view of the voluminous nature of the references and the likelihood that these references are available to the Examiner, copies of references _____ are not enclosed.

☐ In accordance with 37 C.F.R. § 1.98(d), copies of the following references listed on the attached PTO Form SB/08A and PTO Form SB/08B, formerly known as PTO Form 1449, are not enclosed herewith because they were previously cited by or submitted to the U.S. Patent and Trademark Office in patent application(s) for which a claim for priority under 35 U.S.C. § 120 has been made in the instant application.

☐ Copies of the references listed on the attached PTO Forms SB/08A and SB/08B were previously cited by or submitted to the U.S. Patent and Trademark Office in parent application Serial No. _____, filed _____.

☐ If any of the foregoing publication is not available to the Examiner, Applicant will endeavor to supply a copy at the Examiner's request.

Statement under 37 C.F.R. § 1.97(e)

☐ The undersigned attorney hereby states that each item of information contained in the Information Disclosure Statement was cited in a communication from a foreign patent office in a counterpart application not more than three months prior to the filing of the Information Disclosure Statement.

Statement under 37 C.F.R. § 1.704(d)

☐ The undersigned attorney hereby states that each item of information contained in the Information Disclosure Statement was cited in a communication from a foreign patent office in a counterpart application and the communication was not received by any individual designated in § 1.56(c) more than 30 days prior to the filing of the Information Disclosure Statement.

Fees

☐ No Fee is owed by the applicant(s).

☒ The Information Disclosure Statement Fee of **$260.00** under 37 C.F.R. § 1.17(p) is authorized to be charged to Deposit Account No. 50-3111.

Method of Payment of Fees

☐ Attached is a check in the amount of $_____.

☐ Charge Deposit Account No. 50-3111 in the amount of _____.

☒ via EFS Web - **$260.00**.

☐ Please charge any deficiency or credit any overpayment to Deposit Account 50-3111.

☐ No fee or Statement is required under 37 C.F.R. § 1.97(b).

☐ There are no listed references which are not in the English language.

Dated: November 11, 2021

Respectfully submitted,

/Aaron S. Lukas, Reg.# 59,443/

Aaron S. Lukas, Ph.D.
Registration No. 59,443
Customer No. 171052
COZEN O'CONNOR
1200 Nineteenth Street, N.W.
Washington, DC 20036
Email: alukas@cozen.com
Telephone: (202) 912-4823

Enclosures

Electronic Patent Application Fee Transmittal

Application Number:	16262902
Filing Date:	30-Jan-2019
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Filer:	Aaron Scott Lukas/Valerie Dotson
Attorney Docket Number:	INLX.0198U2/484799

Filed as Large Entity

Filing Fees for Utility under 35 USC 111(a)

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Pages:				
Claims:				
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
Extension-of-Time:				

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Miscellaneous:				
SUBMISSION- INFORMATION DISCLOSURE STMT	1806	1	260	260
		Total in USD ($)		**260**

Electronic Acknowledgement Receipt

EFS ID:	44259428
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	171052
Filer:	Aaron Scott Lukas/Valerie Dotson
Filer Authorized By:	Aaron Scott Lukas
Attorney Docket Number:	INLX.0198U2/484799
Receipt Date:	11-NOV-2021
Filing Date:	30-JAN-2019
Time Stamp:	13:23:02
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	CARD
Payment was successfully received in RAM	$260
RAM confirmation Number	E2021AAD24034104
Deposit Account	
Authorized User	

The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows:

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1		INLX_0198U2_484799_Response_to_Office_Action.pdf	217551 e4d928d66a65adc4cc2eb56db3707569e13e4296	yes	7

	Multipart Description/PDF files in .zip description		
	Document Description	**Start**	**End**
	Amendment/Req. Reconsideration-After Non-Final Reject	1	1
	Claims	2	4
	Applicant Arguments/Remarks Made in an Amendment	5	7

Warnings:

Information:

2	Information Disclosure Statement (IDS) Form (SB08)	INLX_0198U2_484799_IDS.pdf	1042485 baa871e25a4a614fbe192efeaa9604b5619de686	no	5

Warnings:

Information:

3	Foreign Reference	INLX_0198U2_484799_CN10-5312044_Chinese_English.pdf	1311451 036b9f4e64dd363b80c9e17452f6ecd2502d445d	no	21

Warnings:

Information:

4	Foreign Reference	INLX_0198U2_484799_WO2004078336A2.pdf	1869422 556e268cd43d35dff8b8a059c039cd5e95ec1b6f	no	13

Warnings:

Information:

5	Foreign Reference	INLX_0198U2_484799_WO2013077904A1.pdf	2682427 e8e0b7542ca7e201a24bbb112438795153 6410e8	no	19

Warnings:					
Information:					
6	Foreign Reference	INLX_0198U2_484799_WO2013106065A1.pdf	4072337 fc242119b5d04d3eed632f491146189fa1b323de	no	26
Warnings:					
Information:					
7	Foreign Reference	INLX_0198U2_CN10-4926600_Chinese_English_Translation.pdf	1508680 c8fd0c4ceedbf338a04f8f5bccf7e5745213264e	no	22
Warnings:					
Information:					
8	Foreign Reference	INLX_0198U2_JP2008-007463_Japanese_English_Translation.pdf	1526183 ccc9cf8dd6414c7f7e811cd507095f410b036c98	no	27
Warnings:					
Information:					
9	Non Patent Literature	INLX_0198U2_484799_InternationalSearchReport_WrittenOpinion-2019-04-19.pdf	794106 84657d14a935fb22b21709348737dd12a2893979	no	9
Warnings:					
Information:					
10	Non Patent Literature	INLX_0198U2_484799_NPL_ASTMD6866-05_StandardTestMethods.pdf	2087908 5223ea7375b0a5b61c6c6be0d9ea8717e61e267c	no	14
Warnings:					
Information:					
11	Non Patent Literature	INLX_0198U2_484799_NPL_MetabolicEngineering.pdf	1290630 921f82d2e495bb9e1a84d105761325915526c625	no	11
Warnings:					
Information:					
12	Transmittal Letter	INLX_0198U2_484799_IDS_Transmittal.pdf	188077 83e284182c3fc3d0d8d94261e956d383afda6fb9	no	4
Warnings:					
Information:					

13	Fee Worksheet (SB06)	fee-info.pdf	38410 f257c402e3c0b3a64d73aa9204fecbe28e1fc821	no	2

Warnings:	
Information:	
Total Files Size (in bytes):	18629667

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

PTO/SB/06 (09-11)
Approved for use through 1/31/2014. OMB 0651-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

PATENT APPLICATION FEE DETERMINATION RECORD Substitute for Form PTO-875	Application or Docket Number 16/262,902	Filing Date 01/30/2019	☐ To be Mailed

ENTITY: ☑ LARGE ☐ SMALL ☐ MICRO

APPLICATION AS FILED - PART I

FOR	(Column 1) NUMBER FILED	(Column 2) NUMBER EXTRA		RATE ($)	FEE ($)
☐ BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A		N/A	
☐ SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A		N/A	
☐ EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A		N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	minus 20 =	*		x $100 =	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	minus 3 =	*		x $460 =	
☐ APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $310 ($155 for small entity) for each additional 50 sheets or fraction thereof. See 35 U.S.C. 41(a)(1)(G) and 37 CFR 1.16(s).				
☐ MULTIPLE DEPENDENT CLAIM PRESENT (37 CFR 1.16(j))					
* If the difference in column 1 is less than zero, enter "0" in column 2.				TOTAL	

APPLICATION AS AMENDED - PART II

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT	11/11/2021	CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	* 23	Minus	** 23	= 0		x $100 =	0
	Independent (37 CFR 1.16(h))	* 2	Minus	*** 3	= 0		x $480 =	0
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	0

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT		CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	*	Minus	**	=		x $0 =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=		x $0 =	
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	

* If the entry in column 1 is less than the entry in column 2, write "0" in column 3.	LIE
** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 20, enter "20".	/YOLANDA CHADWICK/
*** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 3, enter "3".	
The "Highest Number Previously Paid For" (Total or Independent) is the highest number found in the appropriate box in column 1.	

This collection of information is required by 37 CFR 1.16. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	INLX.0198U2/484799	1028

171052 7590 12/01/2021

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

EXAMINER
HOLLOMAN, NANNETTE

ART UNIT	PAPER NUMBER
1612	

NOTIFICATION DATE	DELIVERY MODE
12/01/2021	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

nyipdocketing@cozen.com
vdotson@cozen.com

Office Action Summary	Application No. 16/262,902	Applicant(s) Burgo et al.	
	Examiner NANNETTE HOLLOMAN	Art Unit 1612	AIA (FITF) Status Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTHS FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

1)☑ Responsive to communication(s) filed on 11/11/2021.

☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____.

2a)☑ This action is **FINAL.** 2b) ☐ This action is non-final.

3)☐ An election was made by the applicant in response to a restriction requirement set forth during the interview on _____; the restriction requirement and election have been incorporated into this action.

4)☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims*

5) ☑ Claim(s) 42-55 and 72-80 is/are pending in the application.

5a) Of the above claim(s) _____ is/are withdrawn from consideration.

6) ☐ Claim(s) _____ is/are allowed.

7) ☑ Claim(s) 42-55 and 72-80 is/are rejected.

8) ☐ Claim(s) _____ is/are objected to.

9) ☐ Claim(s) _____ are subject to restriction and/or election requirement

* If any claims have been determined allowable, you may be eligible to benefit from the **Patent Prosecution Highway** program at a participating intellectual property office for the corresponding application. For more information, please see http://www.uspto.gov/patents/init_events/pph/index.jsp or send an inquiry to **PPHfeedback@uspto.gov.**

Application Papers

10)☐ The specification is objected to by the Examiner.

11)☐ The drawing(s) filed on _____ is/are: a)☐ accepted or b)☐ objected to by the Examiner.

Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).

Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).

Priority under 35 U.S.C. § 119

12)☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

Certified copies:

a)☐ All b)☐ Some** c)☐ None of the:

1.☐ Certified copies of the priority documents have been received.

2.☐ Certified copies of the priority documents have been received in Application No. _____.

3.☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

** See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

1) ☐ Notice of References Cited (PTO-892)

2) ☑ Information Disclosure Statement(s) (PTO/SB/08a and/or PTO/SB/08b) Paper No(s)/Mail Date _____.

3) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date _____.

4) ☐ Other: _____.

DETAILED ACTION

DETAILED ACTION

Notice of Pre-AIA or AIA Status

The present application, filed on or after March 16, 2013, is being examined under the first inventor to file provisions of the AIA.

Applicants' arguments, filed 11/11/2021, have been fully considered. Rejections and/or objections not reiterated from previous office actions are hereby withdrawn. The following rejections and/or objections are either reiterated or newly applied. They constitute the complete set presently being applied to the instant application.

The text of those sections of Title 35, U.S. Code not included in this action can be found in a prior Office Action.

Claim Rejections - 35 USC § 103

Claims 42-44, 47-55 and 72-80 stand rejected under 35 U.S.C. 103 as being unpatentable over Schmaus et al. (US Patent Pub. 2005/0222276) in view of Wang et al. (Green Chem., 2012, 14, pp. 758-765, previously disclosed). This rejection is maintained.

Rejection

Schmaus et al. disclose the use of mixtures of two, three or more 1,2-alkanediols with chain links of 5 to 10 as antimicrobial active compounds (ABSTRACT). Schmaus et al. disclose the use of a mixture

of 1,2-hexanediol and 1,2-octandediol; herein the proportion of each individual diol should be in the range of 1 to 99% (m/m) ([0032] and [0033]). Schmaus et al. disclose the mixtures ate used for the preservation of perishable products such, as cosmetics, pharmaceutical products and foods ([0034]).

Schmaus et al. differs from the instant claims insofar as they do not disclose wherein the 1,2-alkanediols are bio-1,2-alkanediols.

Wang et al. disclose due to the diminishing fossil fuel reservoirs together with the ever deteriorating environment associated with the worldwide fossil fuel based industry, it is imperative to make use of alternative feedstock for production of chemicals and fuels (p.758, Introduction).

It would have been obvious to one of ordinary skill in the art before the effective filing date of the claimed invention to have used "bio" based 1,2-alkanediols in the formulation of Schmaus motivated by the desire to preserve the environment as taught by Wang.

Arguments

Applicant's Arguments

Applicant argues Wang's processes are limited to the production of 1,2-alkanediols with carbon numbers in the range of 2-6. Conversely, the current claims require bio-1,2-alkanediols having a carbon chain length of 7 to 20 carbon atoms. Applicant argues Wang does not teach or suggest ways that a person of ordinary skill in the art could have modified Wang's process to create longer chain 1, 2-alkanediols. Applicant's arguments have been fully considered but they are not persuasive.

Examiner's Response

Wang was used to show why one of ordinary skill in the art would have been motivated to use bio based diols not the process of making said compounds. As previously disclosed, Wang et al. teaches

due to the diminishing fossil fuel reservoirs together with the ever deteriorating environment associated with the worldwide fossil fuel based industry, it is imperative to make use of alternative feedstock for production of chemicals and fuels (p.758, Introduction). Therefore, providing the motivation to use bio-based diols in the formulations of the primary reference.

Claim 42-55 and 72-80 stand rejected under 35 U.S.C. 103 as being unpatentable over Lutz (US Patent Pub. 2012/0282207, previously disclosed) in view of Wang et al. (Green Chem., 2012, 14, pp 758-765, previously disclosed). This rejection is maintained.

Rejection

Lutz discloses an antimicrobial compositions and a product comprising a preservative effective amount of the said composition (Abstract). Lutz discloses household; fabric care and pharmaceutical products ([0043] and claim 25). Lutz discloses the composition comprises caprylyl glycol, which meets the limitation of 1,2-octanediol, pentylene glycol, hexylene glycol, and any combination thereof (reference claim 7). Lutz discloses the composition further comprises an antimicrobial mixture (reference claim 1).

Lutz differs from the instant claims insofar as it does not disclose the 1,2-alkanediol as "bio".

Wang et al. disclose due to the diminishing fossil fuel reservoirs together with the ever-deteriorating environment associated with the worldwide fossil-fuel-based industry, it is imperative to make use of alternative feedstock for the production of chemicals and fuels (p. 758, Introduction).

It would have been obvious to one of ordinary skill in the art before the effective filing date of the claimed invention to have used "bio" based 1,2-alkanediols in the formulation of Lutz motivated by the desire to preserve the environment as taught by Wang.

Arguments

Applicant's Arguments

Applicant argues Wang's processes are limited to the production of 1,2-alkanediols with carbon numbers in the range of 2-6. Conversely, the current claims require bio-1,2-alkanediols having a carbon chain length of 7 to 20 carbon atoms. Applicant argues Wang does not teach or suggest ways that a person of ordinary skill in the art could have modified Wang's process to create longer chain 1, 2-alkanediols. Applicant's arguments have been fully considered but they are not persuasive.

Examiner's Response

Wang was used to show why one of ordinary skill in the art would have been motivated to use bio based diols not the process of making said compounds. As previously disclosed, Wang et al. teaches due to the diminishing fossil fuel reservoirs together with the ever deteriorating environment associated with the worldwide fossil fuel based industry, it is imperative to make use of alternative feedstock for production of chemicals and fuels (p.758, Introduction). Therefore, providing the motivation to use bio-based diols in the formulations of the primary reference.

Conclusion

THIS ACTION IS MADE FINAL. Applicant is reminded of the extension of time policy as set forth in 37 CFR 1.136(a).

A shortened statutory period for reply to this final action is set to expire THREE MONTHS from the mailing date of this action. In the event a first reply is filed within TWO MONTHS of the mailing date of this final action and the advisory action is not mailed until after the end of the THREE-MONTH shortened statutory period, then the shortened statutory period will expire on the date the advisory action is mailed, and any extension fee pursuant to 37 CFR 1.136(a) will be calculated from the mailing date of the advisory action. In no event, however, will the statutory period for reply expire later than SIX MONTHS from the mailing date of this final action.

/NANNETTE HOLLOMAN/
Primary Examiner, Art Unit 1612

EAST Search History

EAST Search History (Prior Art)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
L1	4	("20050069997" | "7385092" | "8450543" | "9732295").PN.	US-PGPUB; USPAT	OR	OFF	2021/11/26 11:34
L2	4	("20050069997" | "7385092" | "8450543" | "9732295").PN.	US-PGPUB; USPAT	OR	OFF	2021/11/26 11:35
S1	937	((("BURGO") near3 ("Rocco")) OR (("WRIGHT") near3 ("Michael")) OR (("MOSSER") near3 ("Gary")) OR (("FEVOLA") near3 ("Michael"))).INV.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	OFF	2020/07/07 19:12
S2	378	(bio with alkene) bioalkene	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/07/07 19:19
S3	6,577	regioselectivity	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/09 14:30
S4	13	regioselectivity same alkanediol	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/09 14:30
S5	7	regioselectivity same (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/09 14:47
S6	1	("7385092").PN.	USPAT; USOCR	OR	OFF	2020/09/09 14:53
S7	2	wo-2016007196-$.did.	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	OFF	2020/09/11 11:19
S8	6	alkanediol same (household and pharmaceutical and cosmetic)	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:25

S9	3	(household and pharmaceutical and cosmetic) same (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26
S10	336	(household and pharmaceutical and cosmetic) and (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26
S11	6	((household and pharmaceutical and cosmetic) and (octanediol or (caprylyl adj glycol))).clm.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26
S12	11	((household and pharmaceutical and cosmetic) and (octanediol or ((caprylyl or hexylene or decylene) adj glycol) or alkanediol)).clm.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:47
S13	946	((("BURGO") near3 ("Rocco")) OR (("WRIGHT") near3 ("Michael")) OR (("MOSSER") near3 ("Gary")) OR (("FEVOLA") near3 ("Michael"))).INV.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	OFF	2020/09/11 19:14
S14	3	S13 and alkanediol	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/09/11 19:18
S15	7	(("8912373") or ("9856461") or ("4234752") or ("20050065389") or ("20030065233") or ("20060161025") or ("20140066666")).PN.	US-PGPUB; USPAT; USOCR	OR	OFF	2020/09/13 13:37
S16	42,261	decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/02/27 23:31
S17	13,209	S16 and (preservative or pharmaceutical or cosmetic)	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/02/27 23:32
S18	1,488	S17 and "bio"	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/02/27 23:32

S19	67	"bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol)))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/04/10 19:56
S20	549,665	((A01N47/44 OR A01N37/36 OR A01N59/16 OR A01N31/02 OR A01N25/34 OR A01N59/00 OR A01N61/00 OR A01N25/00 OR A01N2300/00 OR A01N25/10 OR C07C31/20 OR C07C1/24 OR C07C11/02 OR C07C29/48 OR C07C29/80 OR C07C263/12 OR C07C265/14 OR C07C2521/04 OR C07C11/107 OR C07C29/04 OR C07C247/24 OR C07C11/167 OR C07C11/18 OR C07C2527/054 OR C07C2527/167 OR C07C2527/173 OR C07C2531/02 OR C07C65/21 OR A61K8/345 OR A61K2300/00 OR A61K2800/31 OR A61K8/60 OR A61K2800/412 OR A61K8/11 OR A61K8/732 OR A61K8/738 OR A61K31/197 OR A61K8/44 OR A61K8/73 OR A61K2800/48 OR A61K8/25 OR A61K8/34 OR A61K8/585 OR A61K8/92 OR A61K8/9789 OR A61K9/0014 OR A61K2800/30 OR A61K31/198 OR A61K36/28 OR A61K36/534 OR A61K36/67 OR A61K36/886 OR A61K45/06 OR A61K47/10 OR A61K8/24 OR A61K8/362 OR A61K8/37 OR A61K8/41 OR A61K8/42 OR A61K8/4973 OR A61K8/498 OR A61K8/891 OR A61K9/06 OR A61K2800/10 OR A61K2800/28 OR A61K2800/524 OR A61K2800/56 OR A61K2800/87 OR A61K2800/874 OR A61K31/327 OR A61K47/22 OR A61K47/44 OR A61K47/46 OR A61K8/0216 OR A61K8/046 OR A61K8/19 OR A61K8/20 OR A61K8/23 OR A61K8/26 OR A61K8/31 OR A61K8/64 OR A61K8/678 OR A61K8/735 OR A61K8/8111 OR A61K8/8147 OR A61K8/893 OR A61K8/895 OR A61K8/922 OR A61K8/9794 OR A61K9/107 OR A61K2035/115 OR A61K2800/43 OR A61K2800/522 OR A61K2800/592 OR A61K2800/5922 OR A61K2800/594 OR A61K2800/596 OR A61K2800/624 OR A61K2800/805 OR A61K31/192 OR A61K31/355 OR	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/04/10 19:57

		A61K31/60 OR A61K33/30 OR A61K35/745 OR A61K35/747 OR A61K36/185 OR A61K36/53 OR A61K47/02 OR A61K47/06 OR A61K47/12 OR A61K47/14 OR A61K47/183 OR A61K47/24 OR A61K47/34 OR A61K47/38 OR A61K49/0006 OR A61K49/0008 OR A61K8/06 OR A61K8/062 OR A61K8/28 OR A61K8/29 OR A61K8/342 OR A61K8/347 OR A61K8/365 OR A61K8/368 OR A61K8/375 OR A61K8/416 OR A61K8/445 OR A61K8/463 OR A61K8/49 OR A61K8/4926 OR A61K8/63 OR A61K8/66 OR A61K8/675 OR A61K8/8152 OR A61K8/8194 OR A61K8/927 OR A61K8/965 OR A61K8/97 OR A61K9/08 OR A61Q15/00 OR A61Q19/00 OR A61Q13/00 OR A61Q11/00 OR A61Q5/00 OR A61Q19/007 OR A61Q19/08 OR A61Q19/02 OR A61Q17/005 OR A61Q17/04 OR A61Q19/10 OR A61Q5/02 OR A61Q7/00).CPC.)				
S21	42	S20 and ("bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/04/10 19:57
S24	2	(("9242226") or ("20050222276")).PN.	US-PGPUB; USPAT; USOCR	OR	OFF	2021/08/14 21:51

EAST Search History (Interference)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
S22	125,400	((A01N47/44 OR A01N37/36 OR A01N59/16 OR A01N31/02 OR A01N25/34 OR A01N59/00 OR A01N61/00 OR A01N25/00 OR A01N2300/00 OR A01N25/10 OR C07C31/20 OR C07C1/24 OR C07C11/02 OR C07C29/48 OR C07C29/80 OR C07C263/12 OR C07C265/14 OR C07C2521/04 OR C07C11/107 OR C07C29/04 OR C07C247/24 OR C07C11/167 OR C07C11/18 OR C07C2527/054 OR C07C2527/167 OR C07C2527/173 OR C07C2531/02 OR C07C65/21 OR A61K8/345 OR A61K2300/00 OR A61K2800/31 OR A61K8/60 OR	USPAT	OR	ON	2021/04/10 19:59

		A61K2800/412 OR A61K8/11 OR A61K8/732 OR A61K8/738 OR A61K31/197 OR A61K8/44 OR A61K8/73 OR A61K2800/48 OR A61K8/25 OR A61K8/34 OR A61K8/585 OR A61K8/92 OR A61K8/9789 OR A61K9/0014 OR A61K2800/30 OR A61K31/198 OR A61K36/28 OR A61K36/534 OR A61K36/67 OR A61K36/886 OR A61K45/06 OR A61K47/10 OR A61K8/24 OR A61K8/362 OR A61K8/37 OR A61K8/41 OR A61K8/42 OR A61K8/4973 OR A61K8/498 OR A61K8/891 OR A61K9/06 OR A61K2800/10 OR A61K2800/28 OR A61K2800/524 OR A61K2800/56 OR A61K2800/87 OR A61K2800/874 OR A61K31/327 OR A61K47/22 OR A61K47/44 OR A61K47/46 OR A61K8/0216 OR A61K8/046 OR A61K8/19 OR A61K8/20 OR A61K8/23 OR A61K8/26 OR A61K8/31 OR A61K8/64 OR A61K8/678 OR A61K8/735 OR A61K8/8111 OR A61K8/8147 OR A61K8/893 OR A61K8/895 OR A61K8/922 OR A61K8/9794 OR A61K9/107 OR A61K2035/115 OR A61K2800/43 OR A61K2800/522 OR A61K2800/592 OR A61K2800/5922 OR A61K2800/594 OR A61K2800/596 OR A61K2800/624 OR A61K2800/805 OR A61K31/192 OR A61K31/355 OR A61K31/60 OR A61K33/30 OR A61K35/745 OR A61K35/747 OR A61K36/185 OR A61K36/53 OR A61K47/02 OR A61K47/06 OR A61K47/12 OR A61K47/14 OR A61K47/183 OR A61K47/24 OR A61K47/34 OR A61K47/38 OR A61K49/0006 OR A61K49/0008 OR A61K8/06 OR A61K8/062 OR A61K8/28 OR A61K8/29 OR A61K8/342 OR A61K8/347 OR A61K8/365 OR A61K8/368 OR A61K8/375 OR A61K8/416 OR A61K8/445 OR A61K8/463 OR A61K8/49 OR A61K8/4926 OR A61K8/63 OR A61K8/66 OR A61K8/675 OR A61K8/8152 OR A61K8/8194 OR A61K8/927 OR A61K8/965 OR A61K8/97 OR A61K9/08 OR A61Q15/00 OR A61Q19/00 OR A61Q13/00 OR A61Q11/00 OR A61Q5/00 OR A61Q19/007 OR A61Q19/08 OR A61Q19/02 OR A61Q17/005 OR A61Q17/04 OR A61Q19/10 OR A61Q5/02 OR A61Q7/00).CPC.)				
S23	10	S22 and ("bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))))	USPAT	OR	ON	2021/04/10 19:59

11/26/2021 11:57:05 AM
C:\Users\nholloman\Documents\EAST\Workspaces\16262902.wsp

Doc code: IDS
Doc description: Information Disclosure Statement (IDS) Filed

PTO/SB/08a (02-18)
Approved for use through 11/30/2020. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Not for submission under 37 CFR 1.99)

Application Number	16262902
Filing Date	2019-01-30
First Named Inventor	BURGO, Rocco V.
Art Unit	1612
Examiner Name	HOLLOMAN, Nannette
Attorney Docket Number	INLX.0198U2/484799

U.S.PATENTS Remove

Examiner Initial*	Cite No	Patent Number	Kind Code[1]	Issue Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear
	1	7385092	B2	2008-06-10	Osterholt, et al.	
	2	8450543	B2	2008-05-13	Peters, et al.	
	3	9732295	B1	2017-08-15	Wright, et al.	

If you wish to add additional U.S. Patent citation information please click the Add button. Add

U.S.PATENT APPLICATION PUBLICATIONS Remove

Examiner Initial*	Cite No	Publication Number	Kind Code[1]	Publication Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear
	1	20050069997	A1	2005-03-31	Adkesson, et al.	

If you wish to add additional U.S. Published Application citation information please click the Add button. Add

FOREIGN PATENT DOCUMENTS Remove

Examiner Initial*	Cite No	Foreign Document Number[3]	Country Code[2] i	Kind Code[4]	Publication Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear	T[5]
	1	104926600	CN	A	2015-09-23	XUEYI, Hu, et al.		X

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

INFORMATION DISCLOSURE STATEMENT BY APPLICANT (Not for submission under 37 CFR 1.99)		
	Application Number	16262902
	Filing Date	2019-01-30
	First Named Inventor	BURGO, Rocco V.
	Art Unit	1612
	Examiner Name	HOLLOMAN, Nannette
	Attorney Docket Number	INLX.0198U2/484799

	2	105312044	CN	A	2016-02-10	ZHAOSHENG, Jin, et al.		☒
	3	2008007463	JP	A	2008-01-17	TATSURO, Yamamoto, et al.		☒
	4	2004078336	WO	A2	2004-09-16	ZIEHE, Holgar, et al.		☐
	5	2013077904	WO	A1	2013-05-30	WRIGHT, Michael E.		☐
	6	2013106065	WO	A1	2013-07-18	WRIGHT, Michael E.		☐

If you wish to add additional Foreign Patent Document citation information please click the Add button Add

NON-PATENT LITERATURE DOCUMENTS Remove

Examiner Initials*	Cite No	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc), date, pages(s), volume-issue number(s), publisher, city and/or country where published.	T5
	1	ASTMD6866-05, Standard Test Methods for Determining the Biobased Content of Natural Range Materials Using Radiocarbon and Isotape Ratio Mass Spectrometry Analysis, Mar. 1. 2005	
	2	Lu, Metabolic Engineering 44:77 (Nov 1, 2017)	
	3	International Search Report and Written Opinion of International Application No. PCT1US19/15928 dated Apr. 19, 2019	

If you wish to add additional non-patent literature document citation information please click the Add button Add

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

INFORMATION DISCLOSURE STATEMENT BY APPLICANT (Not for submission under 37 CFR 1.99)		
	Application Number	16262902
	Filing Date	2019-01-30
	First Named Inventor	BURGO, Rocco V.
	Art Unit	1612
	Examiner Name	HOLLOMAN, Nannette
	Attorney Docket Number	INLX.0198U2/484799

EXAMINER SIGNATURE			
Examiner Signature	/NANNETTE HOLLOMAN/	Date Considered	11/26/2021

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through a citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

[1] See Kind Codes of USPTO Patent Documents at www.USPTO.GOV or MPEP 901.04. [2] Enter office that issued the document, by the two-letter code (WIPO Standard ST.3). [3] For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. [4] Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. [5] Applicant is to place a check mark here if English language translation is attached.

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Not for submission under 37 CFR 1.99)

Application Number	16262902
Filing Date	2019-01-30
First Named Inventor	BURGO, Rocco V.
Art Unit	1612
Examiner Name	HOLLOMAN, Nannette
Attorney Docket Number	INLX.0198U2/484799

CERTIFICATION STATEMENT

Please see 37 CFR 1.97 and 1.98 to make the appropriate selection(s):

That each item of information contained in the information disclosure statement was first cited in any communication from a foreign patent office in a counterpart foreign application not more than three months prior to the filing of the information disclosure statement. See 37 CFR 1.97(e)(1).

OR

☐ That no item of information contained in the information disclosure statement was cited in a communication from a foreign patent office in a counterpart foreign application, and, to the knowledge of the person signing the certification after making reasonable inquiry, no item of information contained in the information disclosure statement was known to any individual designated in 37 CFR 1.56(c) more than three months prior to the filing of the information disclosure statement. See 37 CFR 1.97(e)(2).

See attached certification statement.

The fee set forth in 37 CFR 1.17 (p) has been submitted herewith.

☒ A certification statement is not submitted herewith.

SIGNATURE

A signature of the applicant or representative is required in accordance with CFR 1.33, 10.18. Please see CFR 1.4(d) for the form of the signature.

Signature	/Aaron S. Lukas, Reg.# 59,443/	Date (YYYY-MM-DD)	2021-11-11
Name/Print	Aaron S. Lukas, Ph.D.	Registration Number	59,443

This collection of information is required by 37 CFR 1.97 and 1.98. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 1 hour to complete, including gathering, preparing and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

Privacy Act Statement

The Privacy Act of 1974 (P.L. 93-579) requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C. 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether the Freedom of Information Act requires disclosure of these record s.

2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.

3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.

4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).

5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.

6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).

7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.

8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspections or an issued patent.

9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor: **Rocco V. Burgo**	Attorney Docket No: **INLX.0198U2/484799**
Application No.: **16/262,902**	Confirmation No.: **1028**
Filing Date: **January 20, 2019**	Art Unit: **1612**
Title: ***Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same***	Examiner: **HOLLOMAN, Nannette**

FILED VIA EFS

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Summary of Discussion Points for Examiner Interview

In advance of the Examiner interview to be held Wednesday, January 19, 2022, from 1:00 to 1:30 PM (Eastern), Applicants submit the following summary of points for discussion:

The Current Office Action & Cited Prior Art

Concerning the primary prior art references cited in the Final Office Action dated December 1, 2021 (the "Office Action"), the Office readily admits that neither Schmaus (US Patent Pub. No. 2005/0222276) nor Lutz (US Patent Pub. No. 2012/0282207), discloses a bio-1,2-alkanediol in which the carbon atoms of the bio-1,2-alkanediol are from a plant source and are not petrochemically derived, as required by the pending claims. *See* Office Action at 3-4. To overcome these admitted deficiencies, the Office alleges that Wang (*Green Chem 14*:758-65 (2012)) would have motivated a person of ordinary skill in the art to "use bio-based diols in the formulations" of Schmaus and/or Lutz.

Thus, the Office has rejected claims 42-44, 47-55, and 72-80 under 35 U.S.C. § 103 as allegedly obvious over Schmaus in view of Wang, and has rejected claims 42-55 and 72-80 under 35 U.S.C. § 103 as allegedly obvious over Lutz in view of Wang. Office Action at 2-5.

The Office's Response to Applicants' Arguments

The Office notes that Wang was not cited for its teaching of processes to make bio-based C1-C6 diols from cellulosic feedstocks, but rather the desirability of bio-based diols of any chain length.

Subject Matter to be Discussed During the Interview

First, Applicants wish to note that the prior art must enable a person of ordinary skill in the art to make the claimed compositions. *See* MPEP §§ 2121.01-2121.02. Here, the Office has not alleged that either Schmaus or Lutz discloses bio-based C7 to C20 diols, let alone how to make such compounds. And Applicants do not believe there is any dispute that Wang's disclosure is limited to the preparation of C1 to C6 diols. As a result, even if a person of ordinary skill in the art viewing Schmaus or Lutz in view of Wang would have been motivated to prepare compositions that contain bio-1,2-alkanediols, the prior art of record fails to provide any disclosure how to do so. And the mere disclosure of petrochemical-based 1,2-alkanediols cannot render the present claims obvious because that is the very problem that Applicants solved. *See, e.g.,* ¶ [0028] of the as-filed specification.

Second, Applicants note that the claims (in very similar form) were previously found allowable over Lutz in view of Wang. *See* Notices of Allowability dated April 14, 2021 and May 13, 2021. Applicants wish to note that the Office's earlier patentability analysis should similarly apply to the prior art combination of Schmaus in view of Wang.

Third, and finally, Applicants wish to discuss why a person of ordinary skill in the art viewing the prior art of record would not have reasonably expected success in preparing the claimed compositions. *See* MPEP § 2143.02, I-III.

Possible Claim Amendments

An amendment to pending claim 42 to delete the term "first" from the claim is proposed in order to clarify the claim. However, in the event that upon reconsideration of the present obviousness rejection the Office believes that any claim amendment(s) would place this application in condition for allowance, then Applicants look forward to discussing such amendments during tomorrow's interview.

CONCLUSION

Applicants and their undersigned representative look forward to discussing the prior art of record and the current Office Action with the Office.

Respectfully submitted,

Date: January 18, 2022

/Aaron Lukas, Reg.# 59,443/

Aaron S. Lukas, Ph.D.
Reg. No. 59,443
COZEN O'CONNOR
1200 19th Street N.W.
Washington, D.C. 20036
Tel: (202) 912-4823
Fax: (305) 358-3309

Electronic Acknowledgement Receipt

EFS ID:	44763088
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	171052
Filer:	Aaron Scott Lukas/Valerie Dotson
Filer Authorized By:	Aaron Scott Lukas
Attorney Docket Number:	INLX.0198U2/484799
Receipt Date:	18-JAN-2022
Filing Date:	30-JAN-2019
Time Stamp:	10:53:11
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Miscellaneous Incoming Letter	INLX_0198U2_484799_Summary_of_Discussion_Points.pdf	192159 1c84960e58c0190fcf1f7726645b684d36683606	no	3
Warnings:					

Information:	
Total Files Size (in bytes):	192159

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	INLX.0198U2/484799	1028

171052 7590 01/25/2022

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

EXAMINER
HOLLOMAN, NANNETTE

ART UNIT	PAPER NUMBER
1612	

NOTIFICATION DATE	DELIVERY MODE
01/25/2022	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

nyipdocketing@cozen.com
vdotson@cozen.com

Applicant-Initiated Interview Summary	**Application No.** 16/262,902	**Applicant(s)** Burgo et al.		
	Examiner NANNETTE HOLLOMAN	**Art Unit** 1612	**AIA (First Inventor to File) Status** Yes	**Page** 1 of 1

All Participants (applicant, applicants representative, PTO personnel)	**Title**	**Type**
NANNETTE HOLLOMAN	Primary Examiner	Telephonic
AARON LUKAS	Attorney of Record	
MICHAEL FEVOLA	Inventor	

Date of Interview: 19 January 2022

Issues Discussed:

35 U.S.C. 103

Applicant argued the prior art does not teach how to make a C7-C20 bio-1,2-alkanediol. The examiner recommended that the claims be further amended to either add the regioselectivity or the process of preparing the compound since the claims are directed to a composition. .

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Applicant is reminded that a complete written statement as to the substance of the interview must be made of record in the application file. It is the applicants responsibility to provide the written statement, unless the interview was initiated by the Examiner and the Examiner has indicated that a written summary will be provided. See MPEP 713.04
Please further see:
MPEP 713.04
Title 37 Code of Federal Regulations (CFR) § 1.133 Interviews, paragraph (b)
37 CFR § 1.2 Business to be transacted in writing

Applicant recordation instructions: The formal written reply to the last Office action must include the substance of the interview. (See MPEP section 713.04). If a reply to the last Office action has already been filed, applicant is given a non-extendable period of the longer of one month or thirty days from this interview date, or the mailing date of this interview summary form, whichever is later, to file a statement of the substance of the interview.

Examiner recordation instructions: Examiners must summarize the substance of any interview of record. A complete and proper recordation of the substance of an interview should include the items listed in MPEP 713.04 for complete and proper recordation including the identification of the general thrust of each argument or issue discussed, a general indication of any other pertinent matters discussed regarding patentability and the general results or outcome of the interview, to include an indication as to whether or not agreement was reached on the issues raised.

Doc Code: A.NE.AFCP
Document Description: After Final Consideration Pilot Program Request

PTO/SB/434 (05-13)

CERTIFICATION AND REQUEST FOR CONSIDERATION UNDER THE AFTER FINAL CONSIDERATION PILOT PROGRAM 2.0

Practitioner Docket No.:	Application No.:	Filing Date:
INLX.0198U2/484799	16/262,902	January 20, 2019

First Named Inventor:	Title:
Rocco V. Burgo	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same

APPLICANT HEREBY CERTIFIES THE FOLLOWING AND REQUESTS CONSIDERATION UNDER THE AFTER FINAL CONSIDERATION PILOT PROGRAM 2.0 (AFCP 2.0) OF THE ACCOMPANYING RESPONSE UNDER 37 CFR 1.116.

1. The above-identified application is (i) an original utility, plant, or design nonprovisional application filed under 35 U.S.C. 111(a) [a continuing application (*e.g.*, a continuation or divisional application) is filed under 35 U.S.C. 111(a) and is eligible under (i)], or (ii) an international application that has entered the national stage in compliance with 35 U.S.C. 371(c).
2. The above-identified application contains an outstanding final rejection.
3. Submitted herewith is a response under 37 CFR 1.116 to the outstanding final rejection. The response includes an amendment to at least one independent claim, and the amendment does not broaden the scope of the independent claim in any aspect.
4. This certification and request for consideration under AFCP 2.0 is the only AFCP 2.0 certification and request filed in response to the outstanding final rejection.
5. Applicant is willing and available to participate in any interview requested by the examiner concerning the present response.
6. This certification and request is being filed electronically using the Office's electronic filing system (EFS-Web).
7. Any fees that would be necessary consistent with current practice concerning responses after final rejection under 37 CFR 1.116, *e.g.*, extension of time fees, are being concurrently filed herewith. [There is no additional fee required to request consideration under AFCP 2.0.]
8. By filing this certification and request, applicant acknowledges the following:
    - Reissue applications and reexamination proceedings are not eligible to participate in AFCP 2.0.
    - The examiner will verify that the AFCP 2.0 submission is compliant, *i.e.*, that the requirements of the program have been met (see items 1 to 7 above). For compliant submissions:
        - The examiner will review the response under 37 CFR 1.116 to determine if additional search and/or consideration (i) is necessitated by the amendment and (ii) could be completed within the time allotted under AFCP 2.0. If additional search and/or consideration is required but cannot be completed within the allotted time, the examiner will process the submission consistent with current practice concerning responses after final rejection under 37 CFR 1.116, *e.g.*, by mailing an advisory action.
        - If the examiner determines that the amendment does not necessitate additional search and/or consideration, or if the examiner determines that additional search and/or consideration is required and could be completed within the allotted time, then the examiner will consider whether the amendment places the application in condition for allowance (after completing the additional search and/or consideration, if required). If the examiner determines that the amendment does not place the application in condition for allowance, then the examiner will contact the applicant and request an interview.
            - The interview will be conducted by the examiner, and if the examiner does not have negotiation authority, a primary examiner and/or supervisory patent examiner will also participate.
            - If the applicant declines the interview, or if the interview cannot be scheduled within ten (10) calendar days from the date that the examiner first contacts the applicant, then the examiner will proceed consistent with current practice concerning responses after final rejection under 37 CFR 1.116.

Signature	Date
/Aaron S. Lukas, Reg.# 59,443/	February 1, 2022
Name (Print/Typed) Aaron S. Lukas, Ph.D	Practitioner Registration No. 59,443

***Note**: This form must be signed in accordance with 37 CFR 1.33. See 37 CFR 1.4(d) for signature requirements and certifications. Submit multiple forms if more than one signature is required, see below*.*

☐ * Total of _____ forms are submitted.

Privacy Act Statement

The **Privacy Act of 1974 (P.L. 93-579)** requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether disclosure of these records is required by the Freedom of Information Act.
2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.
3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.
4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).
5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.
6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).
7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (*i.e.*, GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.
8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspection or an issued patent.
9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor: **Rocco V. Burgo**	Attorney Docket No: **INLX.0198U2/484799**
Application No.: **16/262,902**	Confirmation No.: **1028**
Filing Date: **January 20, 2019**	Art Unit: **1612**
Title: ***Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same***	Examiner: **HOLLOMAN, Nannette**

VIA EFS

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

RESPONSE TO FINAL OFFICE ACTION UNDER 37 C.F.R. § 1.116

In response to the Final Office Action mailed December 1, 2021, Form PTO/SB/434 requesting a review under AFCP 2.0 is submitted herewith. Applicants' counsel of record is willing and available to participate in any interview initiated by the Examiner concerning the response filed with the AFCP submission. Applicants respectfully request reconsideration of this application in view of the following claim amendments and remarks:

A marked-up **Listing of the Claims** begin on page 2.

Remarks begin on page 5.

LISTING OF THE CLAIMS

This listing of claims will replace all prior versions, and listings, of claims in the application.

What is Claimed is:

1-41. (Canceled)

42. (Currently Amended) A preservative composition, comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of 7 to 20 carbon atoms, wherein the carbon atoms of the [[first]] bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived, and wherein the bio-1,2-alkanediol is prepared from a bio-1-alkene having 7 to 20 carbon atoms.

43. (Previously Presented) A personal care composition comprising the preservative composition of claim 42.

44. (Previously Presented) The personal care composition of claim 43, wherein the personal care composition is a hair care composition, an oral care composition, a skin care composition or a cosmetic composition.

45. (Previously Presented) The personal care composition of claim 43, wherein the personal care composition is a household product.

46. (Previously Presented) The personal care composition of claim 45, wherein the household product is a fabric care product or a cleaning product.

47. (Previously Presented) The personal care composition of claim 43, wherein the bio-1,2-alkanediol is selected from the group consisting of: bio-1,2-heptanediol, bio-1,2-octanediol, bio-1,2-decanediol, bio-1,2-dodecanediol, and combinations thereof.

48. (Previously Presented) The personal care composition of claim 43, wherein the personal care composition is a pharmaceutical composition, a vitamin composition, or a health care composition.

49. (Previously Presented) The personal care composition of claim 43, wherein the preservative composition is present in a concentration of about 0.1% to about 10%.

50. (Previously Presented) The personal care composition of claim 43, further comprising a bio-compound selected from the group consisting of: a bio-organic acid, bio-hydroxamic acid, a bio-alkylhydroxamic acid, a bio-terpenoid compound having 5 to 20 carbon

atoms, bio-1,3-propanediol, bio-1,2-butanediol, bio-1,2-pentanediol, and combinations thereof.

51. (Previously Presented) The personal care composition of claim 50, wherein the preservative composition is present in a concentration of about 0.1% to about 10%.
52. (Previously Presented) The preservative composition of claim 42, wherein the composition further comprises at least one other bio-compound different from the bio-1,2-alkanediol.
53. (Previously Presented) The preservative composition of claim 52, further comprising an antimicrobial compound different from the-bio-1,2-alkanediol.
54. (Previously Presented) The preservative composition of claim 42, comprising about 75% to about 99% of the bio-1,2-alkanediol.
55. (Previously Presented) The preservative composition of claim 42, wherein the bio-1,2-alkanediol comprises about 60% to about 99% bio-1,2-octanediol.

56-71. (Canceled).

72. (Currently Amended) A preservative composition comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of 7 to 14 carbon atoms, wherein the carbon atoms of the bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived, and wherein the bio-1,2-alkanediol is prepared from a bio-alkene having 7 to 14 carbon atoms.
73. (Previously Presented) The preservative composition of claim 72, wherein the bio-1,2-alkanediol is selected from the group consisting of: bio-1,2-heptanediol, bio-1,2-octanediol, bio-1,2-decanediol, bio-1,2-dodecanediol, and combinations thereof.
74. (Previously Presented) The preservative composition of claim 72, wherein the bio-1,2-alkanediol comprises about 60% to about 99% of bio-1,2-octanediol.
75. (Previously Presented) The preservative composition of claim 72, wherein the bio-1,2-alkanediol comprises about 75% to about 99% bio-1,2-octanediol.
76. (Previously Presented) The preservative composition of claim 72, further comprising a bio-compound selected from the group consisting of: a bio-organic acid, bio-hydroxamic acid, a bio-alkylhydroxamic acid, a bio-terpenoid compound having 5 to 20 carbon atoms, bio-1,3-propanediol, bio-1,2-butanediol, bio-1,2-pentanediol, bio-1,2-hexanediol, and combinations thereof.

77. (Previously Presented) A personal care composition comprising the preservative composition of claim 72 in a concentration of about 0.1% to about 10%.

78. (Previously Presented) The personal care composition of claim 77, wherein the preservative composition is present in a concentration of about 0.3% to about 2%.

79. (Previously Presented) The personal care composition of claim 78, wherein the preservative composition comprises about 60% to about 99% of bio-1,2-octanediol.

80. (Previously Presented) The personal care composition of claim 78, wherein the preservative composition comprises about 75% to about 99% of bio-1,2-octanediol.

81-87. (Canceled).

88. (New) A preservative composition, comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of 7 to 20 carbon atoms, wherein the carbon atoms of the bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived, and wherein the preservative composition comprises less than 20% of a corresponding bio-2,3-alkanediol having the same carbon chain length as the bio-1,2-alkanediol.

89. (New) The preservative composition of claim 88, wherein the bio-1,2-alkanediol has a carbon chain length of 7 to 14 carbon atoms.

90. (New) The preservative composition of claim 89, comprising less than about 8% of the corresponding bio-2,3-alkanediol having the same carbon chain length as the bio-1,2-alkanediol.

91. (New) The preservative composition of claim 90, comprising less than about 5% of the corresponding bio-2,3-alkanediol having the same carbon chain length as the bio-1,2-alkanediol.

REMARKS

Application and Claims Status

Upon entry of the foregoing claim amendments, claims 42-55, 72-80, and 88-91 are pending. Claims 1-41, 56-71, and 81-87 were previously cancelled.

Claims 42 and 72 are sought to be amended, and claims 88-91 are sought to be added. Written support for the amendments to claims 42 and 72, and new claims 88-91, can be found in the as-filed specification at, *inter alia*, ¶¶ [0029]-[0030], [0037], [0061], [0065]-[0067], [0070], [0091]-[0092], [0126]-[0130], FIG. 1, and the Abstract.

Claims 42-55 and 72-80 are rejected. In view of the following remarks, Applicants respectfully submit that the pending rejections have been traversed and respectfully request favorable reconsideration and allowance of the pending claims.

Request to Participate in AFCP 2.0

Applicants respectfully request to participate in the After Final Consideration Pilot Program 2.0. Applicants submit that all requirements are met including the additional filed transmittal form PTO/SB/434 and a response including an amendment to at least one independent claim that does not broaden the scope of the claims. Applicants, and Applicants' counsel of record, are willing and available to participate in any interview initiated by the Examiner concerning the accompanying response.

Examiner Interview

Applicants thank the Examiner for taking time to speak with their undersigned counsel of record and Mike Fevola, Ph.D. on Wednesday, January 19, 2022. During the interview, the prior art rejection was discussed, as well as possible claim amendments that may place this application into allowable form. Aspects of the discussion are incorporated into the above claim amendments and following remarks.

Patentability of the Claims

The Rejections For Obviousness Under 35 U.S.C. § 103 Should be Withdrawn

Claims 42-44, 47-55, and 72-80 were rejected under 35 U.S.C. § 103 as allegedly obvious over Schmaus (US Patent Pub. No. 2005/0222276) in view of Wang (*Green Chem.*, 14:758-65 (2012). Final Office Action at 2-4.

Claims 42-55 and 72-80 were rejected under 35 U.S.C. § 103 as allegedly obvious over Lutz (US Patent Pub. No. 2012/0282207) in view of Wang. Final Office Action at 4-5.

Based on the foregoing claim amendments, Applicants respectfully submit that claims 42-55 and 72-80 are in condition for allowance. Specifically, during the interview of January 19, 2022, the Examiner expressed that claim amendments reflective of the process by which the bio-1,2-alkanediol compounds are prepared should place the claims in condition for allowance. Independent claims 42 and 72 have been amended to require "wherein the bio-1,2-alkanediol is prepared from a bio-1-alkene having 7 to 20 carbon atoms," and "wherein the bio-1,2-alkanediol is prepared from a bio-1-alkene having 7 to 14 carbon atoms," respectively. As such, claims 42 and 72, and claims 43-55 and 73-80 that depend therefrom, all recite the process by which the claimed bio-1,2-alkanediol compositions are prepared.

As noted in Applicants' Response dated November 11, 2021, neither Schmaus nor Lutz teach or suggest how to prepare any bio-1,2-alkanediols, let alone bio-1,2-alkanediols having a chain length of 7 to 20 carbon atoms, let alone through a bio-1-alkene intermediate. Wang does not remedy this deficiency, as Wang's disclosure is limited to methods of using cellulosic feedstocks to prepare bio-C_2-C_6 alkanediols,. *See* Wang at 761, Table 1. As such, a person of ordinary skill in the art following Schmaus or Lutz in view of Wang would have no way to make any 1,2-alkanediols through a bio-1-alkene intermediate. For these reasons, Applicants respectfully request reconsideration and withdrawal of the rejections over Schmaus in view of Wang and Lutz in view of Wang.

New Claims 88-91 Are Similarly Non-Obvious Over the Prior Art of Record

Claims 88-91 require, *inter alia*, "[a] preservative composition, comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of 7 to 20 carbon atoms, . . . wherein the preservative composition comprises less than 20% of a corresponding bio-2,3-alkanediol having the same carbon chain length as the bio-1,2-alkanediol." As discussed in the

as-filed specification, the disclosed process for preparing bio-1,2-alkanediols utilizes a bio-1-alkene intermediate having a regioselectivity of at least 80%. *See, e.g.,* ¶¶ [0065]-[0067]. This regioselectivity importantly minimizes the formation of the corresponding bio-2,3-alkanediol impurity. *See, e.g.,* ¶¶ [0126]-[0128] (Examples 5-7, describing a bio-1-alkene having a regioselectivity of 96%, which then provides a bio-1,2-alkanediol purity of 98.7%). That is, the conversion of the bio-1-alkene to the corresponding claimed bio-1,2-alkanediol occurs with near unity yield, and therefore, the bio-1-alkene regioselectivity is largely responsible for providing the purity of the resulting claimed bio-1,2-alkanediol composition. Thus, Applicants respectfully submit that new claims 88-91, which require "less than 20% of a corresponding bio-2,3-alkanediol having the same carbon chain length as the bio-1,2-alkanediol," are reflective of the process disclosed in the instant application.

For these reasons, Applicants respectfully submit that new claims 88-91 are in condition for allowance.

CONCLUSION

In view of the foregoing claim amendments and remarks, this Amendment After Final Rejection places this application in condition for allowance, and therefore, its entry is proper under 37 C.F.R. § 1.116. Applicants respectfully submit that the pending claims are in condition for allowance and Notice of Allowance is earnestly solicited. The Examiner is invited to contact the undersigned attorney with any questions, comments or suggestions relating to the referenced patent application.

This response is being submitted on January 31, 2022. In the event the Commissioner determines that any fees are required for any reason, the Commissioner is hereby authorized to deduct said fee from Deposit Account No.: 50-3111/Matter No.: 484799.000.

Respectfully submitted,

Date: February 1, 2022

/Aaron Lukas, Reg.# 59,443/

Aaron S. Lukas, Ph.D.
Reg. No. 59,443
COZEN O'CONNOR
1200 Nineteenth Street N.W.
Washington, D.C. 20036
Tel: (202) 912-4823

Fax: (202) 861-1905

Electronic Acknowledgement Receipt

EFS ID:	44887434
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	171052
Filer:	Aaron Scott Lukas/Elizabeth George
Filer Authorized By:	Aaron Scott Lukas
Attorney Docket Number:	INLX.0198U2/484799
Receipt Date:	01-FEB-2022
Filing Date:	30-JAN-2019
Time Stamp:	13:02:02
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	After Final Consideration Program Request	198US2_2022_1_31_PTO_SB_434_AFCP.pdf	248889 307964eacc61ca9a5a1b3eae3c7e5a9f8252e88f	no	2
Warnings:					

Information:					
2		198US2_2022_Response_to_Final_OA.pdf	225984 895dc8ccaa292a1c168409a55d8784606e6cf608	yes	8

	Multipart Description/PDF files in .zip description		
	Document Description	**Start**	**End**
	Response After Final Action	1	1
	Claims	2	4
	Applicant Arguments/Remarks Made in an Amendment	5	8

Warnings:	
Information:	
Total Files Size (in bytes):	474873

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	INLX.0198U2/484799	1028

171052 7590 03/10/2022

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

EXAMINER
HOLLOMAN, NANNETTE

ART UNIT	PAPER NUMBER
1612	

NOTIFICATION DATE	DELIVERY MODE
03/10/2022	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

nyipdocketing@cozen.com
vdotson@cozen.com

Advisory Action Before the Filing of an Appeal Brief	Application No. 16/262,902	Applicant(s) Burgo et al.	
	Examiner NANNETTE HOLLOMAN	Art Unit 1612	AIA (FITF) Status Yes

--The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

THE REPLY FILED 01 February 2022 FAILS TO PLACE THIS APPLICATION IN CONDITION FOR ALLOWANCE.

NO NOTICE OF APPEAL FILED

1. ☑ The reply was filed after a final rejection. No Notice of Appeal has been filed. To avoid abandonment of this application, applicant must timely file one of the following replies: (1) an amendment, affidavit, or other evidence, which places the application in condition for allowance; (2) a Notice of Appeal (with appeal fee) in compliance with 37 CFR 41.31; or (3) a Request for Continued Examination (RCE) in compliance with 37 CFR 1.114 if this is a utility or plant application. Note that RCEs are not permitted in design applications. The reply must be filed within one of the following time periods:
   a) ☐ The period for reply expires ____ months from the mailing date of the final rejection.
   b) ☑ The period for reply expires on: (1) the mailing date of this Advisory Action; or (2) the date set forth in the final rejection, whichever is later. In no event, however, will the statutory period for reply expire later than SIX MONTHS from the mailing date of the final rejection.
   c) ☐ A prior Advisory Action was mailed more than 3 months after the mailing date of the final rejection in response to a first after-final reply filed within 2 months of the mailing date of the final rejection.The current period for reply expires ____ months from the mailing date of *the prior Advisory Action* or SIX MONTHS from the mailing date of the final rejection, whichever is earlier.
   Examiner Note: If box 1 is checked, check either box (a), (b) or (c). ONLY CHECK BOX (b) WHEN THIS ADVISORY ACTION IS THE FIRST RESPONSE TO APPLICANTS FIRST AFTER-FINAL REPLY WHICH WAS FILED WITHIN TWO MONTHS OF THE FINAL REJECTION. ONLY CHECK BOX (c) IN THE LIMITED SITUATION SET FORTH UNDER BOX (c). See MPEP 706.07(f).

Extensions of time may be obtained under 37 CFR 1.136(a). The date on which the petition under 37 CFR 1.136(a) and the appropriate extension fee have been filed is the date for purposes of determining the period of extension and the corresponding amount of the fee. The appropriate extension fee under 37 CFR 1.17(a) is calculated from: (1) the expiration date of the shortened statutory period for reply originally set in the final Office action; or (2) as set forth in (b) or (c) above, if checked. Any reply received by the Office later than three months after the mailing date of the final rejection, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

NOTICE OF APPEAL

2. ☐ The Notice of Appeal was filed on _____. A brief in compliance with 37 CFR 41.37 must be filed within two months of the date of filing the Notice of Appeal (37 CFR 41.37(a)), or any extension thereof (37 CFR 41.37(e)), to avoid dismissal of the appeal. Since a Notice of Appeal has been filed, any reply must be filed within the time period set forth in 37CFR 41.37(a).

AMENDMENTS

3. ☑ The proposed amendments filed after a final rejection, but prior to the date of filing a brief, will not be entered because
   a) ☑ They raise new issues that would require further consideration and/or search (see NOTE below);
   b) ☐ They raise the issue of new matter (see NOTE below);
   c) ☑ They are not deemed to place the application in better form for appeal by materially reducing or simplifying the issues for appeal; and/or
   d) ☑ They present additional claims without canceling a corresponding number of finally rejected claims.
   NOTE: See Continuation Sheet (See 37 CFR 1.116 and 41.33(a)).
4. ☐ The amendments are not in compliance with 37 CFR 1.121. See attached Notice of Non-Compliant Amendment (PTOL-324).
5. ☐ Applicant's reply has overcome the following rejection(s): _____
6. ☐ Newly proposed or amended claim(s) _____ would be allowable if submitted in a separate, timely filed amendment canceling the non-allowable claim(s).
7. ☐ For purposes of appeal, the proposed amendment(s):(a) ☐ will not be entered, or (b) ☐ will be entered, and an explanation of how the new or amended claims would be rejected is provided below or appended.

AFFIDAVIT OR OTHER EVIDENCE

8. ☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____
9. ☐ The affidavit or other evidence filed after final action, but before or on the date of filing a Notice of Appeal will not be entered because applicant failed to provide a showing of good and sufficient reasons why the affidavit or other evidence is necessary and was not earlier presented. See 37 CFR 1.116(e).
10. ☐ The affidavit or other evidence filed after the date of filing the Notice of Appeal, but prior to the date of filing a brief, will not be entered because the affidavit or other evidence failed to overcome all rejections under appeal and/or appellant fails to provide a showing of good and sufficient reasons why it is necessary and was not earlier presented. See 37 CFR 41.33(d)(1).
11. ☐ The affidavit or other evidence is entered. An explanation of the status of the claims after entry is below or attached.

REQUEST FOR RECONSIDERATION/OTHER

12. ☑ The request for reconsideration has been considered but does NOT place the application in condition for allowance because: See Continuation Sheet.
13. ☐ Note the attached Information *Disclosure Statement*(s). (PTO/SB/08) Paper No(s). _____
14. ☑ Other: PTO 2323 attached.

STATUS OF CLAIMS

15. The status of the claim(s) is (or will be) as follows:
    Claim(s) allowed:_____.
    Claim(s) objected to:_____.
    Claim(s) rejected:42-55 and 72-80.
    Claim(s) withdrawn from consideration:_____.

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Continuation of 3. NOTE: Applicant has proposed to add new claims 88-91 and amend claims 42 and 72 to recite the limitation "and wherein the bio-1,2-alkanediol is prepared from a bio-1-alkene having 7 to 20 carbon atoms". This limitation was not previously considered in regard to the compositions of claims 42 and 72 nor the dependent claims, and further analysis would be required to determine whether limitations would have been obvious. Additionally a further search would also have to be made to determine the state of the art with regard to this issue.

In regard to the After Final Consideration Program, additional search and/or consideration is necessitated by the amendment that cannot be completed in the alloted time. Therefore, the submission will be processed with current practice concerning responses after final rejection.

Continuation of REQUEST FOR RECONSIDERATION/OTHER 12. The request for reconsideration has been considered but does NOT place the application in condition for allowance because: Applicant's arguments that the newly amended claims are patentable over the prior art references are moot at this time due to non-entry of the proposed amendment.

AFCP 2.0 Decision

Application No.	Applicant(s)	
16/262,902	Burgo et al.	
Examiner	**Art Unit**	**AIA (FITF) Status**
NANNETTE HOLLOMAN	1612	Yes

This is in response to the After Final Consideration Pilot request filed 01 February 2022.

1. **Improper Request** – The AFCP 2.0 request is improper for the following reason(s) and the after final amendment submitted with the request will be treated under pre-pilot procedure.

   ☐ An AFCP 2.0 request form PTO/SB/434 (or equivalent document) was not submitted.

   ☐ A non-broadening amendment to at least one independent claim was not submitted.

   ☐ The request is not the first proper AFCP 2.0 request submitted in response to the most recent final rejection.

   ☐ Other: _____

2. **Proper Request**

   A. After final amendment submitted with the request will not be treated under AFCP 2.0.
   The after final amendment cannot be reviewed and a search conducted within the guidelines of the pilot program.

   ☑ The after final amendment will be treated under pre-pilot procedure.

   B. Updated search and/or completed additional consideration.
   The examiner performed an updated search and/or completed additional consideration of the after final amendment within the time authorized for the pilot program. The result(s) of the updated search and/or completed additional consideration are:

   ☐ 1. All of the rejections in the most recent final Office action are overcome and a Notice of Allowance is issued herewith.

   ☐ 2. The after final amendment would not overcome all of the rejections in the most recent final Office action . See attached interview summary for further details.

   ☐ 3. The after final amendment was reviewed, and it raises a new issue(s). See attached interview summary for further details.

   ☐ 4. The after final amendment raises new issues, but would overcome all of the rejections in the most recent final Office action. A decision on determining allowability could not be made within the guidelines of the pilot. See attached interview summary for further details, including any newly discovered prior art.

   ☐ 5. Other: _____

   Examiner Note: Please attach an interview summary when necessary as described above.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor: **Rocco V. Burgo**	Attorney Docket No: **INLX.0198U2/484799**
Application No.: **16/262,902**	Confirmation No.: **1028**
Filing Date: **January 20, 2019**	Art Unit: **1612**
Title: ***Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same***	Examiner: **HOLLOMAN, Nannette**

VIA EFS

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

RESPONSE TO FINAL OFFICE ACTION UNDER 37 C.F.R. § 1.116

In response to the Final Office Action mailed December 1, 2021, Form PTO/SB/434 requesting a review under AFCP 2.0 is submitted herewith. Applicants' counsel of record is willing and available to participate in any interview initiated by the Examiner concerning the response filed with the AFCP submission. Applicants respectfully request reconsideration of this application in view of the following claim amendments and remarks:

A marked-up **Listing of the Claims** begin on page 2.

Remarks begin on page 5.

PTO/SB/06 (09-11)
Approved for use through 1/31/2014. OMB 0651-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

PATENT APPLICATION FEE DETERMINATION RECORD Substitute for Form PTO-875	Application or Docket Number 16/262,902	Filing Date 01/30/2019	☐ To be Mailed

ENTITY: ☑ LARGE ☐ SMALL ☐ MICRO

APPLICATION AS FILED - PART I

FOR	(Column 1) NUMBER FILED	(Column 2) NUMBER EXTRA		RATE ($)	FEE ($)
☐ BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A		N/A	
☐ SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A		N/A	
☐ EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A		N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	minus 20 =	*		x $100 =	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	minus 3 =	*		x $460 =	
☐ APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $310 ($155 for small entity) for each additional 50 sheets or fraction thereof. See 35 U.S.C. 41(a)(1)(G) and 37 CFR 1.16(s).				
☐ MULTIPLE DEPENDENT CLAIM PRESENT (37 CFR 1.16(j))					
* If the difference in column 1 is less than zero, enter "0" in column 2.				TOTAL	

APPLICATION AS AMENDED - PART II

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT	03/16/2022	CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	* 27	Minus	** 27	= 0		x $100 =	0
	Independent (37 CFR 1.16(h))	* 2	Minus	*** 3	= 0		x $480 =	0
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	0

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT		CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	*	Minus	**	=		x $0 =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=		x $0 =	
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	

* If the entry in column 1 is less than the entry in column 2, write "0" in column 3.	LIE
** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 20, enter "20".	/VICTOR BARLOW/
*** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 3, enter "3".	
The "Highest Number Previously Paid For" (Total or Independent) is the highest number found in the appropriate box in column 1.	

This collection of information is required by 37 CFR 1.16. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

PTO/SB/30EFS (01-22)
Approved for use through 05/31/2024. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

REQUEST FOR CONTINUED EXAMINATION(RCE)TRANSMITTAL
(Submitted Only via EFS-Web)

Application Number	16/262,902	Filing Date	2019-01-30	Docket Number (if applicable)	INLX.0198U2/484799	Art Unit	1612
First Named Inventor	BURGO, Rocco V.			Examiner Name	HOLLOMAN, Nannette		

This is a Request for Continued Examination (RCE) under 37 CFR 1.114 of the above-identified application.
Request for Continued Examination (RCE) practice under 37 CFR 1.114 does not apply to any utility or plant application filed prior to June 8, 1995, or to any design application. The Instruction Sheet for this form is located at WWW.USPTO.GOV

SUBMISSION REQUIRED UNDER 37 CFR 1.114

Note: If the RCE is proper, any previously filed unentered amendments and amendments enclosed with the RCE will be entered in the order in which they were filed unless applicant instructs otherwise. If applicant does not wish to have any previously filed unentered amendment(s) entered, applicant must request non-entry of such amendment(s).

☐ Previously submitted. If a final Office action is outstanding, any amendments filed after the final Office action may be considered as a submission even if this box is not checked.

- ☐ Consider the arguments in the Appeal Brief or Reply Brief previously filed on _____
- ☐ Other _____

☒ Enclosed

- ☒ Amendment/Reply
- ☒ Information Disclosure Statement (IDS)
- ☐ Affidavit(s)/ Declaration(s)
- ☒ Other Extension of Time (1 month)

MISCELLANEOUS

☐ Suspension of action on the above-identified application is requested under 37 CFR 1.103(c) for a period of months _____ (Period of suspension shall not exceed 3 months; Fee under 37 CFR 1.17(i) required)

☐ Other _____

FEES

☒ **The RCE fee under 37 CFR 1.17(e) is required by 37 CFR 1.114 when the RCE is filed.**
The Director is hereby authorized to charge any underpayment of fees, or credit any overpayments, to Deposit Account No 503111

SIGNATURE OF APPLICANT, ATTORNEY, OR AGENT REQUIRED

☒ Patent Practitioner Signature

Applicant Signature

PTO/SB/30EFS (01-22)
Approved for use through 05/31/2024. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

Signature of Registered U.S. Patent Practitioner			
Signature	/kathryn a warner/	Date (YYYY-MM-DD)	2022-03-16
Name	Kathryn A. Warner	Registration Number	59858

This collection of information is required by 37 CFR 1.114. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.11 and 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450.

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

Privacy Act Statement

The Privacy Act of 1974 (P.L. 93-579) requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C. 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether the Freedom of Information Act requires disclosure of these records.

2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.

3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.

4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).

5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.

6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).

7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.

8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspections or an issued patent.

9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

PTO/SB/08a (01-22)
Approved for use through 05/31/2024. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Not for submission under 37 CFR 1.99)

Application Number	16262902
Filing Date	2019-01-30
First Named Inventor	BURGO, Rocco V.
Art Unit	1612
Examiner Name	HOLLOMAN, Nannette
Attorney Docket Number	INLX.0198U2/484799

U.S.PATENTS Remove

Examiner Initial*	Cite No	Patent Number	Kind Code[1]	Issue Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear
	1					

If you wish to add additional U.S. Patent citation information please click the Add button. Add

U.S.PATENT APPLICATION PUBLICATIONS Remove

Examiner Initial*	Cite No	Publication Number	Kind Code[1]	Publication Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear
	1	20090143489	A1	2009-06-04	WINN, Daniel	
	2	20090004122	A1	2009-01-01	MODAK, Shanta, et al.	
	3	20120238788	A1	2012-09-20	WRIGHT, Michael E.	

If you wish to add additional U.S. Published Application citation information please click the Add button. Add

FOREIGN PATENT DOCUMENTS Remove

Examiner Initial*	Cite No	Foreign Document Number[3]	Country Code[2] i	Kind Code[4]	Publication Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear	T[5]
	1	3045161	EP	A1	2016-07-20	SYMRISE AG		

INFORMATION DISCLOSURE STATEMENT BY APPLICANT (Not for submission under 37 CFR 1.99)		
	Application Number	16262902
	Filing Date	2019-01-30
	First Named Inventor	BURGO, Rocco V.
	Art Unit	1612
	Examiner Name	HOLLOMAN, Nannette
	Attorney Docket Number	INLX.0198U2/484799

If you wish to add additional Foreign Patent Document citation information please click the Add button Add

NON-PATENT LITERATURE DOCUMENTS Remove

Examiner Initials*	Cite No	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc), date, pages(s), volume-issue number(s), publisher, city and/or country where published.	T[5]
	1	Supplementary European Search Report from European Application No. EP 19 74 7575; mailing date of February 24, 2022; Paul Campbell (Examiner)	

If you wish to add additional non-patent literature document citation information please click the Add button Add

EXAMINER SIGNATURE

Examiner Signature		Date Considered	

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through a citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

[1] See Kind Codes of USPTO Patent Documents at www.USPTO.GOV or MPEP 901.04. [2] Enter office that issued the document, by the two-letter code (WIPO Standard ST.3). [3] For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. [4] Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. [5] Applicant is to place a check mark here if English language translation is attached.

INFORMATION DISCLOSURE STATEMENT BY APPLICANT (Not for submission under 37 CFR 1.99)		
	Application Number	16262902
	Filing Date	2019-01-30
	First Named Inventor	BURGO, Rocco V.
	Art Unit	1612
	Examiner Name	HOLLOMAN, Nannette
	Attorney Docket Number	INLX.0198U2/484799

CERTIFICATION STATEMENT

Please see 37 CFR 1.97 and 1.98 to make the appropriate selection(s):

☒ That each item of information contained in the information disclosure statement was first cited in any communication from a foreign patent office in a counterpart foreign application not more than three months prior to the filing of the information disclosure statement. See 37 CFR 1.97(e)(1).

OR

☐ That no item of information contained in the information disclosure statement was cited in a communication from a foreign patent office in a counterpart foreign application, and, to the knowledge of the person signing the certification after making reasonable inquiry, no item of information contained in the information disclosure statement was known to any individual designated in 37 CFR 1.56(c) more than three months prior to the filing of the information disclosure statement. See 37 CFR 1.97(e)(2).

See attached certification statement.

The fee set forth in 37 CFR 1.17 (p) has been submitted herewith.

A certification statement is not submitted herewith.

SIGNATURE

A signature of the applicant or representative is required in accordance with CFR 1.33, 10.18. Please see CFR 1.4(d) for the form of the signature.

Signature	/kathryn a warner/	Date (YYYY-MM-DD)	2022-03-16
Name/Print	Kathryn A. Warner	Registration Number	59,858

This collection of information is required by 37 CFR 1.97 and 1.98. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 1 hour to complete, including gathering, preparing and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

Privacy Act Statement

The Privacy Act of 1974 (P.L. 93-579) requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C. 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether the Freedom of Information Act requires disclosure of these record s.

2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.

3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.

4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).

5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.

6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).

7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.

8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspections or an issued patent.

9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) EP 3 045 161 A1

(12) **EUROPEAN PATENT APPLICATION**

(43) Date of publication:
20.07.2016 Bulletin 2016/29

(21) Application number: **15151564.0**

(22) Date of filing: **18.01.2015**

(51) Int Cl.:
A61K 8/34 *(2006.01)* ***A61K 8/06*** *(2006.01)*
A61Q 19/00 *(2006.01)*

(84) Designated Contracting States:
AL AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HR HU IE IS IT LI LT LU LV MC MK MT NL NO PL PT RO RS SE SI SK SM TR
Designated Extension States:
BA ME

(71) Applicant: **Symrise AG**
37603 Holzminden (DE)

(72) Inventors:
- **Lange, Sabine**
37603 Holzminden (DE)
- **Schmaus, Gerhard**
37671 Höxter (DE)

(74) Representative: **Fabry, Bernd**
IP2 Patentanwalts GmbH
Schlossstrasse 523-525
41238 Mönchengladbach (DE)

Remarks:
A request for correction of the title has been filed pursuant to Rule 139 EPC. A decision on the request will be taken during the proceedings before the Examining Division (Guidelines for Examination in the EPO, A-V, 3.).

(54) **Active compositions comprising 1,2-hexanediol and 1,2-octanediol**

(57) Suggested is an active mixture comprising
(a) 1,2-hexanediol and
(b) 1,2-octanediol.

Printed by Jouve, 75001 PARIS (FR)

Description

FIELD OF INVENTION

[0001] The present invention belongs to the area of cosmetics and refers to active mixtures consisting of two selected 1,2-alkanediols, compositions comprising said actives and their specific applications.

STATE OF THE ART

[0002] Emulsions are one of the most important types of formulation for all kind of skin care products with a wide range of applications. Therefore, developing formulations with excellent physical and microbial stability is a major challenge for cosmetic industry.

[0003] Due to legal restrictions or negative press the number of accepted substances for microbial stabilization is limited. Therefore more and more traditional preservatives are replaced totally or partly by multifunctional ingredients with antimicrobial properties.

[0004] Along antimicrobial efficacy physiochemical stability of emulsions is yet another important factor for product development.

[0005] Finally, sensorial profile of the cosmetic product is of utmost importance for acceptance by the customer. Particularly, spreadability, absorption and skin feeling are important parameters contributing to the overall performance of cosmetic formulations after application to skin. The faster the oil bodies of an emulsion are spread on the skin, the better is the perception of the customer. The spreading behaviour of a formulation - and therefore its sensorial profile - is linked to the average particle size of the droplets in the composition. The smaller the droplets are, the faster the spreading is. As a consequence, there is still a need for additives allowing to shift the average particle size distribution to lower values. Also, particle size is an important parameter for emulsion stability: the smaller the droplets of the dispersed phase, the better the physiochemical stability.

[0006] According to international patent application WO 2003 069994 A1 it is known that mixtures of alkane diols of different change lengths have a synergistically enhanced antimicrobial effect in comparison to pure 1,2-alkane diols. However, the document neither teaches nor suggests using diols for modifying the particle size distribution within an emulsion towards smaller droplets.

[0007] Therefore, the object of the present invention has been providing actives for use in cosmetic, pharmaceutical or dermatological emulsions which simultaneously

- provide antimicrobial stability
- shift the average particle size within the compositions towards smaller droplets, and
- improve sensory profile, particularly with regard to spreadability, absorption and skin feeling.

DESCRIPTION OF THE INVENTION

[0008] Object of the present invention is an active mixture comprising

(a) 1,2-hexanediol and

(b) 1,2-octanediol.

[0009] Surprisingly, it has been observed that mixtures of 1,2-hexanediol and 1,2-octanediol, preferably in equal amounts, fully solves the complex problem explained above. In particular the binary mixture shows a synergistic behaviour with respect to antimicrobial activity, decrease of average droplet size and sensory profile when added to a cosmetic, dermatological of pharmaceutical composition.

1,2-ALKANEDIOLS

[0010] 1,2-Alkanediols are well-known cosmetic ingredients obtainable for example by epoxidation of terminal olefins followed by ring cleavage using water. With regard to synergistic activity the best results were obtained with mixtures where the two components (a) and (b) are present in a ratio by weight of from about 25:75 to about 75:25, and more particularly in a ratio by weight of from about 40:60 to about 60:40, even more particular in a ratio by weight of about 50:50.

EMULSIONS

[0011] Another object of the present invention is related to a cosmetic or a pharmaceutical composition comprising a synergistic mixture of 1,2-hexanediol and 1,2-hexanediol as disclosed above. Preferably said composition is a skin care, hair care, sun care or personal care composition.

[0012] Preferably the composition is an emulsion, in particular said emulsion being an oil-in-water (o/w) emulsion, a water-in-oil (w/o) emulsion or a multiple (w/o/w or o/w/o) emulsion.

[0013] Typically, said emulsions exhibit an average particle size, where

- 10 Vol.-% show a diameter of less than 2 μm (preferably less than 0.5 μm),
- 50 Vol.-% show a diameter of less than 5 μm (preferably less than 2 μm) and
- 90 Vol.-% show a diameter of less than 15 μm (preferably less than 12 μm).

COSMETIC OR PERSONAL CARE COMPOSITION

[0014] Another object of the present invention encompasses a cosmetic or personal care composition comprising said binary mixture of 1,2-alkanediols. The cosmetic or personal care composition may represent a skin care, hair care and/or sun care product, such as for example a cosmetic cream, lotion, spray, emulsion, ointment, gel or mouse and the like. Typical examples are skin creams and hair shampoos, antiperspirants and soaps.

[0015] The preparations according to the invention may contain abrasives, anti-acne agents, preservatives, agents against ageing of the skin, anti-cellulitis agents, antidandruff agents, anti-inflammatory agents, irritation-preventing agents, irritation-inhibiting agents, antioxidants, astringents, deodorant agents, perspiration-inhibiting agents, antiseptic agents, anti-statics, binders, buffers, carrier materials, chelating agents, cell stimulants, cleansing agents, care agents, depilatory agents, surface-active substances, deodorizing agents, antiperspirants, softeners, emulsifiers, enzymes, essential oils, fibres, film-forming agents, fixatives, foam-forming agents, foam stabilizers, substances for preventing foaming, foam boosters, gelling agents, gel-forming agents, hair care agents, hair-setting agents, hair-straightening agents, moisture-donating agents, moisturizing substances, moisture-retaining substances, bleaching agents, strengthening agents, stain-removing agents, optically brightening agents, impregnating agents, dirt-repellent agents, friction-reducing agents, lubricants, moisturizing creams, ointments, opacifying agents, plasticizing agents, covering agents, polish, gloss agents, polymers, powders, proteins, re-oiling agents, abrading agents, silicones, skin-soothing agents, skin-cleansing agents, skin care agents, skin-healing agents, skin-lightening agents, skin-protecting agents, skin-softening agents, hair promotion agents, cooling agents, skin-cooling agents, warming agents, skin-warming agents, stabilizers, UV-absorbing agents, UV filters, detergents, fabric conditioning agents, suspending agents, skin-tanning agents, thickeners, vitamins, oils, waxes, fats, phospholipids, saturated fatty acids, mono- or polyunsaturated fatty acids, α-hydroxy acids, polyhydroxyfatty acids, liquefiers, dyestuffs, colour-protecting agents, pigments, anti-corrosives, aromas, flavouring substances, odoriferous substances, polyols, surfactants, electrolytes, organic solvents or silicone derivatives and the like as additional auxiliaries and additives.

SURFACTANS

[0016] Preferred auxiliaries and additives are anionic and/or amphoteric or zwitterionic surfactants. Typical examples of anionic surfactants are soaps, alkyl benzenesulfonates, alkanesulfonates, olefin sulfonates, alkylether sulfonates, glycerol ether sulfonates, methyl ester sulfonates, sulfofatty acids, alkyl sulfates, fatty alcohol ether sulfates, glycerol ether sulfates, fatty acid ether sulfates, hydroxy mixed ether sulfates, monoglyceride (ether) sulfates, fatty acid amide (ether) sulfates, mono- and dialkyl sulfosuccinates, mono- and dialkyl sulfosuccinamates, sulfotriglycerides, amide soaps, ether carboxylic acids and salts thereof, fatty acid isethionates, fatty acid sarcosinates, fatty acid taurides, N-acylamino acids such as, for example, acyl lactylates, acyl tartrates, acyl glutamates and acyl aspartates, alkyl oligoglucoside sulfates, protein fatty acid condensates (particularly wheat-based vegetable products) and alkyl (ether) phosphates. If the anionic surfactants contain polyglycol ether chains, they may have a conventional homolog distribution although they preferably have a narrow-range homolog distribution. Typical examples of amphoteric or zwitterionic surfactants are alkylbetaines, alkylamidobetaines, aminopropionates, aminoglycinates, imidazolinium betaines and sulfobetaines. The surfactants mentioned are all known compounds. Information on their structure and production can be found in relevant synoptic works, cf. for example J. Falbe (ed.), "Surfactants in Consumer Products", Springer Verlag, Berlin, 1987, pages 54 to 124 **or** J. Falbe (ed.), "Katalysatoren, Tenside und Mineralöladditive (Catalysts, Surfactants and Mineral Oil Additives)", Thieme Verlag, Stuttgart, 1978, pages 123-217. The percentage content of surfactants in the preparations may be from 0.1 to 10% by weight and is preferably from 0.5 to 5% by weight, based on the preparation.

OIL BODIES

[0017] Suitable oil bodies, which form constituents of the O/W emulsions, are, for example, Guerbet alcohols based on fatty alcohols having 6 to 18, preferably 8 to 10, carbon atoms, esters of linear C_6-C_{22}-fatty acids with linear or branched C_6-C_{22}-fatty alcohols or esters of branched C_6-C_{13}-carboxylic acids with linear or branched C_6-C_{22}-fatty alcohols, such as, for example, myristyl myristate, myristyl palmitate, myristyl stearate, myristyl isostearate, myristyl oleate, myristyl behenate, myristyl erucate, cetyl myristate, cetyl palmitate, cetyl stearate, cetyl isostearate, cetyl oleate, cetyl behenate, cetyl erucate, stearyl myristate, stearyl palmitate, stearyl stearate, stearyl isostearate, stearyl oleate, stearyl behenate, stearyl erucate, isostearyl myristate, isostearyl palmitate, isostearyl stearate, isostearyl isostearate, isostearyl oleate, isostearyl behenate, isostearyl oleate, oleyl myristate, oleyl palmitate, oleyl stearate, oleyl isostearate, oleyl oleate, oleyl behenate, oleyl erucate, behenyl myristate, behenyl palmitate, behenyl stearate, behenyl isostearate, behenyl oleate, behenyl behenate, behenyl erucate, erucyl myristate, erucyl palmitate, erucyl stearate, erucyl isostearate, erucyl oleate, erucyl behenate and erucyl erucate. Also suitable are esters of linear C_6-C_{22}-fatty acids with branched alcohols, in particular 2-ethylhexanol, esters of C_{18}-C_{38}-alkylhydroxy carboxylic acids with linear or branched C_6-C_{22}-fatty alcohols, in particular Dioctyl Malate, esters of linear and/or branched fatty acids with polyhydric alcohols (such as, for example, propylene glycol, dimerdiol or trimertriol) and/or Guerbet alcohols, triglycerides based on C_6-C_{10}-fatty acids, liquid mono-/di-/triglyceride mixtures based on C_6-C_{18}-fatty acids, esters of C_6-C_{22}-fatty alcohols and/or Guerbet alcohols with aromatic carboxylic acids, in particular benzoic acid, esters of C_2-C_{12}-dicarboxylic acids with linear or branched alcohols having 1 to 22 carbon atoms or polyols having 2 to 10 carbon atoms and 2 to 6 hydroxyl groups, vegetable oils, branched primary alcohols, substituted cyclohexanes, linear and branched C_6-C_{22}-fatty alcohol carbonates, such as, for example, Dicaprylyl Carbonate (Cetiol® CC), Guerbet carbonates, based on fatty alcohols having 6 to 18, preferably 8 to 10, carbon atoms, esters of benzoic acid with linear and/or branched C_6-C_{22}-alcohols (e.g. Fin-solv® TN), linear or branched, symmetrical or asymmetrical dialkyl ethers having 6 to 22 carbon atoms per alkyl group, such as, for example, dicaprylyl ether (Cetiol® OE), ring-opening products of epoxidized fatty acid esters with polyols, silicone oils (cyclomethicones, silicone methicone grades, etc.) and/or aliphatic or naphthenic hydrocarbons, such as, for example, squalane, squalene or dialkylcyclohexanes.

EMULSIFIERS

[0018] Other surfactants may also be added to the preparations as emulsifiers, including for example:

- products of the addition of 2 to 30 mol ethylene oxide and/or 0 to 5 mol propylene oxide onto linear C_{8-22} fatty alcohols, onto C_{12-22} fatty acids and onto alkyl phenols containing 8 to 15 carbon atoms in the alkyl group;
- $C_{12/18}$ fatty acid monoesters and diesters of addition products of 1 to 30 mol ethylene oxide onto glycerol;
- glycerol mono- and diesters and sorbitan mono- and diesters of saturated and unsaturated fatty acids containing 6 to 22 carbon atoms and ethylene oxide addition products thereof;
- addition products of 15 to 60 mol ethylene oxide onto castor oil and/or hydrogenated castor oil;
- polyol esters and, in particular, polyglycerol esters such as, for example, polyglycerol polyricinoleate, polyglycerol poly-12-hydroxystearate or polyglycerol dimerate isostearate. Mixtures of compounds from several of these classes are also suitable;
- addition products of 2 to 15 mol ethylene oxide onto castor oil and/or hydrogenated castor oil;
- partial esters based on linear, branched, unsaturated or saturated $C_{6/22}$ fatty acids, ricinoleic acid and 12-hydroxystearic acid and glycerol, polyglycerol, pentaerythritol, - dipentaerythritol, sugar alcohols (for example sorbitol), alkyl glucosides (for example methyl glucoside, butyl glucoside, lauryl glucoside) and polyglucosides (for example cellulose);
- mono-, di and trialkyl phosphates and mono-, di- and/or tri-PEG-alkyl phosphates and salts thereof;
- wool wax alcohols;
- polysiloxane/polyalkyl polyether copolymers and corresponding derivatives;

- mixed esters of pentaerythritol, fatty acids, citric acid and fatty alcohol and/or mixed esters of C_{6-22} fatty acids, methyl glucose and polyols, preferably glycerol or polyglycerol,
- polyalkylene glycols and
- glycerol carbonate.

[0019] The addition products of ethylene oxide and/or propylene oxide onto fatty alcohols, fatty acids, alkylphenols, glycerol mono- and diesters and sorbitan mono- and diesters of fatty acids or onto castor oil are known commercially available products. They are homologue mixtures of which the average degree of alkoxylation corresponds to the ratio between the quantities of ethylene oxide and/or propylene oxide and substrate with which the addition reaction is carried out. $C_{12/18}$ fatty acid monoesters and diesters of addition products of ethylene oxide onto glycerol are known as lipid layer enhancers for cosmetic formulations. The preferred emulsifiers are described in more detail as follows:

[0020] **Partial glycerides.** Typical examples of suitable partial glycerides are hydroxystearic acid monoglyceride, hydroxystearic acid diglyceride, isostearic acid monoglyceride, isostearic acid diglyceride, oleic acid monoglyceride, oleic acid diglyceride, ricinoleic acid monoglyceride, ricinoleic acid diglyceride, linoleic acid monoglyceride, linoleic acid diglyceride, linolenic acid monoglyceride, linolenic acid diglyceride, erucic acid monoglyceride, erucic acid diglyceride, tartaric acid monoglyceride, tartaric acid diglyceride, citric acid monoglyceride, citric acid diglyceride, malic acid monoglyceride, malic acid diglyceride and technical mixtures thereof which may still contain small quantities of triglyceride from the production process. Addition products of 1 to 30 and preferably 5 to 10 mol ethylene oxide onto the partial glycerides mentioned are also suitable.

[0021] **Sorbitan esters.** Suitable sorbitan esters are sorbitan monoisostearate, sorbitan sesquiisostearate, sorbitan diisostearate, sorbitan triisostearate, sorbitan monooleate, sorbitan sesquioleate, sorbitan dioleate, sorbitan trioleate, sorbitan monoerucate, sorbitan sesquierucate, sorbitan dierucate, sorbitan trierucate, sorbitan monoricinoleate, sorbitan sesquiricinoleate, sorbitan diricinoleate, sorbitan triricinoleate, sorbitan monohydroxystearate, sorbitan sesquihydroxystearate, sorbitan dihydroxystearate, sorbitan trihydroxystearate, sorbitan monotartrate, sorbitan sesquitartrate, sorbitan ditartrate, sorbitan tritartrate, sorbitan monocitrate, sorbitan sesquicitrate, sorbitan dicitrate, sorbitan tricitrate, sorbitan monomaleate, sorbitan sesquimaleate, sorbitan dimaleate, sorbitan trimaleate and technical mixtures thereof. Addition products of 1 to 30 and preferably 5 to 10 mol ethylene oxide onto the sorbitan esters mentioned are also suitable.

[0022] **Polyglycerol esters.** Typical examples of suitable polyglycerol esters are Polyglyceryl-2 Dipolyhydroxystearate (Dehymuls® PGPH), Polyglycerin-3-Diisostearate (Lameform ® TGI), Polyglyceryl-4 Isostearate (Isolan GI 34), Polyglyceryl-3 Oleate, Diisostearoyl Polyglyceryl-3 Diisostearate (Isolan® PDI), Polyglyceryl-3 Methylglucose Distearate (Tego Care 450), Polyglyceryl-3 Beeswax (Cera Bellina®), Polyglyceryl-4 Caprate (Polyglycerol Caprate T2010/90), Polyglyceryl-3 Cetyl Ether (Chimexane®NL), Polyglyceryl-3 Distearate (Cremophor® GS 32) and Polyglyceryl Polyricinoleate (Admul® WOL 1403), Polyglyceryl Dimerate Isostearate and mixtures thereof. Examples of other suitable polyolesters are the mono-, di- and triesters of trimethylol propane or pentaerythritol with lauric acid, cocofatty acid, tallow fatty acid, palmitic acid, stearic acid, oleic acid, behenic acid and the like optionally reacted with 1 to 30 mol ethylene oxide.

[0023] **Anionic emulsifiers.** Typical anionic emulsifiers are aliphatic C_{12-22} fatty acids, such as palmitic acid, stearic acid or behenic acid for example, and C_{12-22} dicarboxylic acids, such as azelaic acid or sebacic acid for example.

[0024] **Amphoteric emulsifiers.** Other suitable emulsifiers are amphboteric or zwitterionic surfactants. Zwitterionic surfactants are surface-active compounds which contain at least one quaternary ammonium group and at least one carboxylate and one sulfonate group in the molecule. Particularly suitable zwitterionic surfactants are the so-called betaines, such as the N-alkyl-N,N-dimethyl ammonium glycinates, for example cocoalkyl dimethyl ammonium glycinate, N-acylaminopropyl-N,N-dimethyl ammonium glycinates, for example cocoacylaminopropyl dimethyl ammonium glycinate, and 2-alkyl-3-carboxymethyl-3-hydroxyethyl imidazolines containing 8 to 18 carbon atoms in the alkyl or acyl group and cocoacylaminoethyl hydroxyethyl carboxymethyl glycinate. The fatty acid amide derivative known under the CTFA name of *Cocamidopropyl Betaine* is particularly preferred. Ampholytic surfactants are also suitable emulsifiers. Ampholytic surfactants are surface-active compounds which, in addition to a $C_{8/18}$ alkyl or acyl group, contain at least one free amino group and at least one -COOH- or -SO_3H- group in the molecule and which are capable of forming inner salts. Examples of suitable ampholytic surfactants are N-alkyl glycines, N-alkyl propionic acids, N-alkylaminobutyric acids, N-alkyliminodipropionic acids, N-hydroxyethyl-N-alkylamidopropyl glycines, N-alkyl taurines, N-alkyl sarcosines, 2-alkylaminopropionic acids and alkylaminoacetic acids containing around 8 to 18 carbon atoms in the alkyl group. Particularly preferred ampholytic surfactants are N-cocoalkylaminopropionate, cocoacylaminoethyl aminopropionate and $C_{12/18}$ acyl sarcosine.

SUPERFATTING AGENTS AND CONSISTENCY FACTORS

[0025] Superfatting agents may be selected from such substances as, for example, lanolin and lecithin and also

polyethoxylated or acylated lanolin and lecithin derivatives, polyol fatty acid esters, monoglycerides and fatty acid alkanolamides, the fatty acid alkanolamides also serving as foam stabilizers.

[0026] The consistency factors mainly used are fatty alcohols or hydroxyfatty alcohols containing 12 to 22 and preferably 16 to 18 carbon atoms and also partial glycerides, fatty acids or hydroxyfatty acids. A combination of these substances with alkyl oligoglucosides and/or fatty acid N-methyl glucamides of the same chain length and/or polyglycerol poly-12-hydroxystearates is preferably used.

THICKENING AGENTS AND RHEOLOGY ADDITIVES

[0027] Suitable thickeners are polymeric thickeners, such as Aerosil® types (hydrophilic silicas), polysaccharides, more especially xanthan gum, guar-guar, agar-agar, alginates and tyloses, carboxymethyl cellulose and hydroxyethyl cellulose, also relatively high molecular weight polyethylene glycol monoesters and diesters of fatty acids, polyacrylates (for example Carbopols® [Goodrich] or Synthalens® [Sigma]), polyacrylamides, polyvinyl alcohol and polyvinyl pyrrolidone, surfactants such as, for example, ethoxylated fatty acid glycerides, esters of fatty acids with polyols, for example pentaerythritol or trimethylol propane, narrow-range fatty alcohol ethoxylates and electrolytes, such as sodium chloride and ammonium chloride.

POLYMERS

[0028] Suitable cationic polymers are, for example, cationic cellulose derivatives such as, for example, the quaternized hydroxyethyl cellulose obtainable from Amerchol under the name of Polymer JR 400®, cationic starch, copolymers of diallyl ammonium salts and acrylamides, quaternized vinyl pyrrolidone/vinyl imidazole polymers such as, for example, Luviquat® (BASF), condensation products of polyglycols and amines, quaternized collagen polypeptides such as, for example, Lauryldimonium Hydroxypropyl Hydrolyzed Collagen (Lamequat® L, Grünau), quaternized wheat polypeptides, polyethyleneimine, cationic silicone polymers such as, for example, amodimethicone, copolymers of adipic acid and dimethylaminohydroxypropyl diethylenetriamine (Cartaretine®, Sandoz), copolymers of acrylic acid with dimethyl diallyl ammonium chloride (Merquat® 550, Chemviron), polyaminopoly-amides and crosslinked water-soluble polymers thereof, cationic chitin derivatives such as, for example, quaternized chitosan, optionally in microcrystalline distribution, condensation products of dihaloalkyls, for example dibromobutane, with bis-dialkylamines, for example bis-dimethylamino-1,3-propane, cationic guar gum such as, for example, Jaguar® CBS, Jaguar C-17, Jaguar C-16 of Celanese, quaternized ammonium salt polymers such as, for example, Mirapol® A-15, Mirapol® AD-1, Mirapol® AZ-1 of Miranol and the various polyquaternium types (for example 6, 7, 32 or 37) which can be found in the market under the tradenames Rheocare® CC or Ultragel® 300.

[0029] Suitable anionic, zwitterionic, amphoteric and nonionic polymers are, for example, vinyl acetate/crotonic acid copolymers, vinyl pyrrolidone/vinyl acrylate copolymers, vinyl acetate/butyl maleate/isobornyl acrylate copolymers, methyl vinylether/maleic anhydride copolymers and esters thereof, uncrosslinked and polyol-crosslinked polyacrylic acids, acrylamidopropyl trimethylammonium chloride/acrylate copolymers, octylacrylamide/methyl methacrylate/tert.-butylaminoethyl methacrylate/2-hydroxypropyl methacrylate copolymers, polyvinyl pyrrolidone, vinyl pyrrolidone/vinyl acetate copolymers, vinyl pyrrolidone/dimethylaminoethyl methacrylate/vinyl caprolactam terpolymers and optionally derivatized cellulose ethers and silicones.

PEARLIZING WAXES

[0030] Suitable pearlising waxes are, for example, alkylene glycol esters, especially ethylene glycol distearate; fatty acid alkanolamides, especially cocofatty acid diethanolamide; partial glycerides, especially stearic acid monoglyceride; esters of polybasic, optionally hydroxysubstituted carboxylic acids with fatty alcohols containing 6 to 22 carbon atoms, especially long-chain esters of tartaric acid; fatty compounds, such as for example fatty alcohols, fatty ketones, fatty aldehydes, fatty ethers and fatty carbonates which contain in all at least 24 carbon atoms, especially laurone and distearylether; fatty acids, such as stearic acid, hydroxystearic acid or behenic acid, ring opening products of olefin epoxides containing 12 to 22 carbon atoms with fatty alcohols containing 12 to 22 carbon atoms and/or polyols containing 2 to 15 carbon atoms and 2 to 10 hydroxyl groups and mixtures thereof.

SILICONES

[0031] Suitable silicone compounds are, for example, dimethyl polysiloxanes, methylphenyl polysiloxanes, cyclic silicones and amino-, fatty acid-, alcohol-, polyether-, epoxy-, fluorine-, glycoside- and/or alkyl-modified silicone compounds which may be both liquid and resin-like at room temperature. Other suitable silicone compounds are simethicones which are mixtures of dimethicones with an average chain length of 200 to 300 dimethylsiloxane units and hydrogenated

silicates. A detailed overview of suitable volatile silicones can be found in Todd et al. in Cosm. Toil. 91, 27 (1976).

WAXES AND STABILIZERS

[0032] Besides natural oils used, waxes may also be present in the preparations, more especially natural waxes such as, for example, candelilla wax, carnauba wax, Japan wax, espartograss wax, cork wax, guaruma wax, rice oil wax, sugar cane wax, ouricury wax, montan wax, beeswax, shellac wax, spermaceti, lanolin (wool wax), uropygial fat, ceresine, ozocerite (earth wax), petrolatum, paraffin waxes and microwaxes; chemically modified waxes (hard waxes) such as, for example, montan ester waxes, sasol waxes, hydrogenated jojoba waxes and synthetic waxes such as, for example, polyalkylene waxes and polyethylene glycol waxes.

[0033] Metal salts of fatty acids such as, for example, magnesium, aluminium and/or zinc stearate or ricinoleate may be used as stabilizers.

PRIMARY SUN PROTECTION FACTORS

[0034] Primary sun protection factors in the context of the invention are, for example, organic substances (light filters) which are liquid or crystalline at room temperature and which are capable of absorbing ultraviolet radiation and of releasing the energy absorbed in the form of longer-wave radiation, for example heat.

[0035] The formulations according to the invention advantageously contain at least one UV-A filter and/or at least one UV-B filter and/or a broadband filter and/or at least one inorganic pigment. Formulations according to the invention preferably contain at least one UV-B filter or a broadband filter, more particularly preferably at least one UV-A filter and at least one UV-B filter.

[0036] Preferred cosmetic compositions, preferably topical formulations according to the present invention comprise one, two, three or more sun protection factors selected from the group consistiung of 4-aminobenzoic acid and derivatives, salicylic acid derivatives, benzophenone derivatives, dibenzoylmethane derivatives, diphenyl acrylates, 3-imidazol-4-yl acrylic acid and esters thereof, benzofuran derivatives, benzylidene malonate derivatives, polymeric UV absorbers containing one or more organosilicon radicals, cinnamic acid derivatives, camphor derivatives, trianilino-s-triazine derivatives, 2-hydroxyphenylbenzotriazole derivatives, phenylbenzimidazole sulfonic acid derivatives and salts thereof, anthranilic acid menthyl esters, benzotriazole derivatives and indole derivatives.

[0037] In addition, it is advantageous to combine compounds of formula (I) with active ingredients which penetrate into the skin and protect the skin cells from inside against sunlight-induced damage and reduce the level of cutaneous matrix metalloproteases. Preferred respective ingredients, so called arylhydrocarbon receptor antagonists, are described in WO 2007/128723, incorporated herein by reference. Preferred is 2-benzylidene-5,6-dimethoxy-3,3-dimethylindan-1-one.

[0038] The UV filters cited below which can be used within the context of the present invention are preferred but naturally are not limiting.

[0039] UV filters which are preferably used are selected from the group consisting of

- p-aminobenzoic acid
- p-aminobenzoic acid ethyl ester (25 mol) ethoxylated (INCI name: PEG-25 PABA)
- p-dimethylaminobenzoic acid-2-ethylhexyl ester
- p-aminobenzoic acid ethyl ester (2 mol) N-propoxylated
- p-aminobenzoic acid glycerol ester
- salicylic acid homomenthyl ester (homosalates) (Neo Heliopan®HMS)
- salicylic acid-2-ethylhexyl ester (Neo Heliopan®OS)
- triethanolamine salicylate
- 4-isopropyl benzyl salicylate
- anthranilic acid menthyl ester (Neo Heliopan®MA)

- diisopropyl cinnamic acid ethyl ester
- p-methoxycinnamic acid-2-ethylhexyl ester (Neo Heliopan®AV)
- diisopropyl cinnamic acid methyl ester
- p-methoxycinnamic acid isoamyl ester (Neo Heliopan®E 1000)
- p-methoxycinnamic acid diethanolamine salt
- p-methoxycinnamic acid isopropyl ester
- 2-phenylbenzimidazole sulfonic acid and salts (Neo Heliopan®Hydro)
- 3-(4'-trimethylammonium) benzylidene bornan-2-one methyl sulfate
- beta-imidazole-4(5)-acrylic acid (urocanic acid)
- 3-(4'-sulfo)benzylidene bornan-2-one and salts
- 3-(4'-methyl benzylidene)-D,L-camphor (Neo Heliopan®MBC)
- 3-benzylidene-D,L-camphor
- N-[(2 and 4)-[2-(oxoborn-3-ylidene) methyl]benzyl] acrylamide polymer
- 4,4'-[(6-[4-(1,1-dimethyl)aminocarbonyl) phenylamino]-1,3,5-triazine-2,4-diyl)diimino]-bis-(benzoic acid-2-ethyl-hexyl ester) (Uvasorb®HEB)
- benzylidene malonate polysiloxane (Parsol®SLX)
- glyceryl ethylhexanoate dimethoxycinnamate
- dipropylene glycol salicylate
- tris(2-ethylhexyl)-4,4',4''-(1,3,5-triazine-2,4,6-triyltriimino)tribenzoate (= 2,4,6-trianilino-(p-carbo-2'-ethylhexyl-1'-oxy)-1,3,5-triazine) (Uvinul®T150).

[0040] Broadband filters which are preferably combined with one or more compounds of formula (I) in a preparation according to the present invention are selected from the group consisting of

- 2-ethylhexyl-2-cyano-3,3-diphenyl acrylate (Neo Heliopan®303)
- ethyl-2-cyano-3,3'-diphenyl acrylate
- 2-hydroxy-4-methoxybenzophenone (Neo Heliopan®BB)
- 2-hydroxy-4-methoxybenzophenone-5-sulfonic acid
- dihydroxy-4-methoxybenzophenone
- 2,4-dihydroxybenzophenone
- tetrahydroxybenzophenone
- 2,2'-dihydroxy-4,4'-dimethoxybenzophenone
- 2-hydroxy-4-n-octoxybenzophenone

- 2-hydroxy-4-methoxy-4'-methyl benzophenone
- sodium hydroxymethoxybenzophenone sulfonate
- disodium-2,2'-dihydroxy-4,4'-dimethoxy-5,5'-disulfobenzophenone
- phenol, 2-(2H-benzotriazol-2-yl)-4-methyl-6-(2-methyl-3(1,3,3,3-tetramethyl-1-(trime-thylsilyl)oxy)disiloxyanyl) propyl) (Mexoryl®XL)
- 2,2'-methylene bis-(6-(2H-benzotriazol-2-yl)-4-1,1,3,3-tetramethylbutyl) phenol) (Tinosorb®M)
- 2,4-bis-[4-(2-ethylhexyloxy)-2-hydroxyphenyl]-1,3,5-triazine
- 2,4-bis-[{(4-(2-ethylhexyloxy)-2-hydroxy}phenyl]-6-(4-methoxyphenyl)-1,3,5-triazine (Tinosorb®S)
- 2,4-bis-[{(4-(3-sulfonato)-2-hydroxypropyloxy)-2-hydroxy}phenyl]-6-(4-methoxyphenyl)-1,3,5-triazine sodium salt
- 2,4-bis-[{(3-(2-propyloxy)-2-hydroxypropyloxy)-2-hydroxy}phenyl]-6-(4-methoxyphenyl)-1,3,5-triazine
- 2,4-bis-[{4-(2-ethylhexyloxy)-2-hydroxy}phenyl]-6-[4-(2-methoxyethyl carbonyl) phenylamino]-1,3,5-triazine
- 2,4-bis-[{4-(3-(2-propyloxy)-2-hydroxypropyloxy)-2-hydroxy}phenyl]-6-[4-(2-ethylcarboxyl) phenylamino]-1,3,5-triazine
- 2,4-bis-[{4-(2-ethylhexyloxy)-2-hydroxy}phenyl]-6-(1-methylpyrrol-2-yl)-1,3,5-triazine
- 2,4-bis-[{4-tris-(trimethylsiloxysilylpropyloxy)-2-hydroxy}phenyl]-6-(4-methoxyphenyl)-1,3,5-triazine
- 2,4-bis-[{4-(2"-methylpropenyloxy)-2-hydroxy}phenyl]-6-(4-methoxyphenyl)-1,3,5-triazine
- 2,4-bis-[{4-(1',1',1',3',5',5',5'-heptamethylsiloxy-2"-methylpropyloxy)-2-hydroxy}phenyl]-6-(4-methoxyphenyl)-1,3,5-triazine.

[0041] The compositions can comprise further typical detergent and cleansing composition ingredients such as UV-A filters filters which are preferably combined with one or more compounds of formula (I) in a preparation according to the present invention are selected from the group consisting of

- 4-isopropyl dibenzoyl methane
- terephthalylidene dibornane sulfonic acid and salts (Mexoryl®SX)
- 4-t-butyl-4'-methoxydibenzoyl methane (avobenzone) / (Neo Heliopan®357)
- phenylene bis-benzimidazyl tetrasulfonic acid disodium salt (Neo Heliopan®AP)
- 2,2'-(1,4-phenylene)-bis-(1H-benzimidazole-4,6-disulfonic acid), monosodium salt
- 2-(4-diethylamino-2-hydroxybenzoyl) benzoic acid hexyl ester (Uvinul® A Plus)
- indanylidene compounds in accordance with DE 100 55 940 A1 (= WO 2002 038537 A1)

[0042] The compositions can comprise further typical detergent and cleansing composition ingredients such as UV filters which are more preferably combined with one or more compounds of formula (I) in a preparation according to the present invention are selected from the group consisting of

- p-aminobenzoic acid
- 3-(4'-trimethylammonium) benzylidene bornan-2-one methyl sulfate

- salicylic acid homomenthyl ester (Neo Heliopan®HMS)
- 2-hydroxy-4-methoxybenzophenone (Neo Heliopan®BB)
- 2-phenylbenzimidazole sulfonic acid (Neo Heliopan®Hydro)
- terephthalylidene dibornane sulfonic acid and salts (Mexoryl®SX)
- 4-tert-butyl-4'-methoxydibenzoyl methane (Neo Heliopan®357)
- 3-(4'-sulfo)benzylidene bornan-2-one and salts
- 2-ethylhexyl-2-cyano-3,3-diphenyl acrylate (Neo Heliopan®303)
- N-[(2 and 4)-[2-(oxoborn-3-ylidene) methyl]benzyl] acrylamide polymer
- p-methoxycinnamic acid-2-ethylhexyl ester (Neo Heliopan®AV)
- p-aminobenzoic acid ethyl ester (25 mol) ethoxylated (INCI name: PEG-25 PABA)
- p-methoxycinnamic acid isoamyl ester (Neo Heliopan®E1000)
- 2,4,6-trianilino-(p-carbo-2'-ethylhexyl-1'-oxy)-1,3,5-triazine (Uvinul®T150)
- phenol, 2-(2H-benzotriazol-2-yl)-4-methyl-6-(2-methyl-3(1,3,3,3-tetramethyl-1-(trime-thylsilyl)oxy)disiloxyanyl) propyl) (Mexoryl®XL)
- 4,4'-[(6-[4-(1,1-dimethyl)aminocarbonyl) phenylamino]-1,3,5-triazine-2,4-diyl)diimino]-bis-(benzoic acid-2-ethylhexyl ester) (Uvasorb HEB)
- 3-(4'-methyl benzylidene)-D,L-camphor (Neo Heliopan®MBC)
- 3-benzylidene camphor
- salicylic acid-2-ethylhexyl ester (Neo Heliopan®OS)
- 4-dimethylaminobenzoic acid-2-ethylhexyl ester (Padimate O)
- hydroxy-4-methoxybenzophenone-5-sulfonic acid and Na salt
- 2,2'-methylene bis-(6-(2H-benzotriazol-2-yl)-4-1,1,3,3-tetramethylbutyl) phenol) (Tinosorb®M)
- phenylene bis-benzimidazyl tetrasulfonic acid disodium salt (Neo Heliopan®AP)
- 2,4-bis-[{(4-(2-ethylhexyloxy)-2-hydroxy}phenyl]-6-(4-methoxyphenyl)-1,3,5-triazine (Tinosorb®S)
- benzylidene malonate polysiloxane (Parsol®SLX)
- menthyl anthranilate (Neo Heliopan®MA)
- 2-(4-diethylamino-2-hydroxybenzoyl) benzoic acid hexyl ester (Uvinul® A Plus)
- indanylidene compounds in accordance with DE 100 55 940 (= WO 02/38537).

[0043] Advantageous primary and also secondary sun protection factors are mentioned in WO 2005 123101 A1. Advantageously, these preparations contain at least one UVA filter and/or at least one UVB filter and/or at least one inorganic pigment. The preparations may be present here in various forms such as are conventionally used for sun protection preparations. Thus, they may be in form of a solution, an emulsion of the water-in-oil type (W/O) or of the oil-

in-water type (O/W) or a multiple emulsion, for example of the water-in-oil-in-water type (W/O/W), a gel, a hydrodispersion, a solid stick or else an aerosol.

[0044] In a further preferred embodiment a formulation according to the invention contains a total amount of sunscreen agents, i.e. in particular UV filters and/or inorganic pigments (UV filtering pigments) so that the formulation according to the invention has a light protection factor of greater than or equal to 2 (preferably greater than or equal to 5). Such formulations according to the invention are particularly suitable for protecting the skin and hair.

SECONDARY SUN PROTECTION FACTORS

[0045] Besides the groups of primary sun protection factors mentioned above, secondary sun protection factors of the antioxidant type may also be used. Secondary sun protection factors of the antioxidant type interrupt the photochemical reaction chain which is initiated when UV rays penetrate into the skin. Typical examples are amino acids (for example glycine, histidine, tyrosine, tryptophane) and derivatives thereof, imidazoles (for example urocanic acid) and derivatives thereof, peptides, such as D,L-carnosine, D-carnosine, L-carnosine and derivatives thereof (for example anserine), carotinoids, carotenes (for example alpha-carotene, beta-carotene, lycopene) and derivatives thereof, chlorogenic acid and derivatives thereof, liponic acid and derivatives thereof (for example dihydroliponic acid), aurothioglucose, propylthiouracil and other thiols (for example thioredoxine, glutathione, cysteine, cystine, cystamine and glycosyl, N-acetyl, methyl, ethyl, propyl, amyl, butyl and lauryl, palmitoyl, oleyl, alpha-linoleyl, cholesteryl and glyceryl esters thereof) and their salts, dilaurylthiodipropionate, distearylthiodipropionate, thiodipropionic acid and derivatives thereof (esters, ethers, peptides, lipids, nucleotides, nucleosides and salts) and sulfoximine compounds (for example butionine sulfoximines, homocysteine sulfoximine, butionine sulfones, penta-, hexa- and hepta-thionine sulfoximine) in very small compatible dosages, also (metal) chelators (for example alpha-hydroxyfatty acids, palmitic acid, phytic acid, lactoferrine), alpha-hydroxy acids (for example citric acid, lactic acid, malic acid), humic acid, bile acid, bile extracts, bilirubin, biliverdin, EDTA, EGTA and derivatives thereof, unsaturated fatty acids and derivatives thereof (for example linoleic acid, oleic acid), folic acid and derivatives thereof, ubiquinone and ubiquinol and derivatives thereof, vitamin C and derivatives thereof (for example ascorbyl palmitate, Mg ascorbyl phosphate, ascorbyl acetate), tocopherols and derivatives (for example vitamin E acetate), vitamin A and derivatives (vitamin A palmitate) and coniferyl benzoate of benzoin resin, rutinic acid and derivatives thereof, glycosyl rutin, ferulic acid, furfurylidene glucitol, carnosine, butyl hydroxytoluene, butyl hydroxyanisole, nordihydroguaiac resin acid, nordihydroguaiaretic acid, trihydroxybutyrophenone, uric acid and derivatives thereof, mannose and derivatives thereof, superoxide dismutase, titanium dioxide (for example dispersions in ethanol), zinc and derivatives thereof (for example ZnO, $ZnSO_4$), selenium and derivatives thereof (for example selenium methionine), stilbenes and derivatives thereof (for example stilbene oxide, trans-stilbene oxide) and derivatives of these active substances suitable for the purposes of the invention (salts, esters, ethers, sugars, nucleotides, nucleosides, peptides and lipids).

[0046] Advantageous inorganic secondary light protection pigments are finely dispersed metal oxides and metal salts which are also mentioned in WO 2005 123101 A1. The total quantity of inorganic pigments, in particular hydrophobic inorganic micro-pigments in the finished cosmetic preparation according to the present invention is advantageously from 0.1 to 30% by weight, preferably 0.5 to 10.0% by weight, in each case based on the total weight of the preparation.

[0047] Also preferred are particulate UV filters or inorganic pigments, which can optionally be hydrophobed, can be used, such as the oxides of titanium (TiO_2), zinc (ZnO), iron (Fe_2O_3), zirconium (ZrO_2), silicon (SiO_2), manganese (e.g. MnO), aluminium (Al_2O_3), cerium (e.g. Ce_2O_3) and/or mixtures thereof.

ACTIVES MODULATING SKIN AND/OR HAIR PIGMENTATION

[0048] Preferred active ingredients for skin and/or hair lightening are selected from the group consisting of: kojic acid (5-hydroxy-2-hydroxymethyl-4-pyranone), kojic acid derivatives, preferably kojic acid dipalmitate, arbutin, ascorbic acid, ascorbic acid derivatives, preferably magnesium ascorbyl phosphate, hydroquinone, hydroquinone derivatives, resorcinol, resorcinol derivatives, preferably 4-alkylresorcinols and 4-(1-phenylethyl)1,3-dihydroxybenzene (phenylethyl resorcinol), cyclohexylcarbamates (preferably one or more cyclohexyl carbamates disclosed in WO 2010/122178 and WO 2010/097480), sulfur-containing molecules, preferably glutathione or cysteine, alpha-hydroxy acids (preferably citric acid, lactic acid, malic acid), salts and esters thereof, N-acetyl tyrosine and derivatives, undecenoyl phenylalanine, gluconic acid, chromone derivatives, preferably aloesin, flavonoids, 1-aminoethyl phosphinic acid, thiourea derivatives, ellagic acid, nicotinamide (niacin-amide), zinc salts, preferably zinc chloride or zinc gluconate, thujaplicin and derivatives, triterpenes, preferably maslinic acid, sterols, preferably ergosterol, benzofuranones, preferably senkyunolide, vinyl guiacol, ethyl guiacol, dionic acids, preferably octodecene dionic acid and/or azelaic acid, inhibitors of nitrogen oxide synthesis, preferably L-nitroarginine and derivatives thereof, 2,7-dinitroindazole or thiocitrulline, metal chelators (preferably alpha-hydroxy fatty acids, phytic acid, humic acid, bile acid, bile extracts, EDTA, EGTA and derivatives thereof), retinoids, soy milk and extract, serine protease inhibitors or lipoic acid or other synthetic or natural active ingredients for

skin and hair lightening, the latter preferably used in the form of an extract from plants, preferably bearberry extract, rice extract, papaya extract, turmeric extract, mulberry extract, bengkoang extract, nutgrass extract, liquorice root extract or constituents concentrated or isolated therefrom, preferably glabridin or licochalcone A, artocarpus extract, extract of rumex and ramulus species, extracts of pine species (pinus), extracts of vitis species or stilbene derivatives isolated or concentrated therefrom, saxifrage extract, scutelleria extract, grape extract and/or microalgae extract, in particular Tetraselmis suecica Extract.

[0049] Preferred skin lighteners as component (b) are kojic acid and phenylethyl resorcinol as tyrosinase inhibitors, beta- and alpha-arbutin, hydroquinone, nicotinamide, dioic acid, Mg ascorbyl phosphate and vitamin C and its derivatives, mulberry extract, Bengkoang extract, papaya extract, turmeric extract, nutgrass extract, licorice extract (containing glycyrrhizin), alpha-hydroxy-acids, 4-alkylresorcinols, 4-hydroxyanisole. These skin lighteners are preferred due to their very good activity, in particular in combination with sclareolide according to the present invention. In addition, said preferred skin lighteners are readily available.

[0050] Advantageous skin and hair tanning active ingredients in this respect are substrates or substrate analogues of tyrosinase such as L-tyrosine, N-acetyl tyrosine, L-DOPA or L-dihydroxyphenylalanine, xanthine alkaloids such as caffeine, theobromine and theophyl-line and derivatives thereof, proopiomelanocortin peptides such as ACTH, alpha-MSH, peptide analogues thereof and other substances which bind to the melanocortin receptor, peptides such as Val-Gly-Val-Ala-Pro-Gly, Lys-Ile- Gly-Arg-Lys or Leu-Ile-Gly-Lys, purines, pyrimidines, folic acid, copper salts such as copper gluconate, chloride or pyrrolidonate, 1,3,4-oxadiazole-2-thiols such as 5-pyrazin-2-yl-1,3,4-oxadiazole-2-thiol, curcumin, zinc diglycinate (Zn(Gly)2), manganese(II) bicarbonate complexes ("pseudocat-alases") as described for example in EP 0 584 178, tetrasubstituted cyclohexene deriva-tives as described for example in WO 2005/032501, isoprenoids as described in WO 2005/102252 and in WO 2006/010661, melanin derivatives such as Melasyn-100 and MelanZe, diacyl glycerols, aliphatic or cyclic diols, psoralens, prostaglandins and ana-logues thereof, activators of adenylate cyclase and compounds which activate the transfer of melanosomes to keratinocytes such as serine proteases or agonists of the PAR-2 receptor, extracts of plants and plant parts of the chrysanthemum species, san-guisorba species, walnut extracts, urucum extracts, rhubarb extracts, microalgae extracts, in particular Isochrysis galbana, trehalose, erythrulose and dihydroxyacetone. Flavonoids which bring about skin and hair tinting or brown-ing (e.g. quercetin, rhamnetin, kaempferol, fisetin, genistein, daidzein, chrysin and api-genin, epicatechin, diosmin and diosmetin, morin, quercitrin, naringenin, hesperidin, phloridzin and phloretin) can also be used.

[0051] The amount of the aforementioned examples of additional active ingredients for the modulation of skin and hair pigmentation (one or more compounds) in the products according to the invention is then preferably 0.00001 to 30 wt.%, preferably 0.0001 to 20 wt.%, particularly preferably 0.001 to 5 wt.%, based on the total weight of the prepa-ration.

ANTI-AGEING ACTIVES

[0052] In the context of the invention, anti-ageing or biogenic agents are, for example antioxidants, matrix-metalloproteinase inhibitors (MMPI), skin moisturizing agents, glycosaminglycan stimulators, anti-inflammatory agents, TRPV1 antagonists and plant extracts.

[0053] **Antioxidants.** Suitable antioxidants encompass amino acids (preferably glycine, histidine, tyrosine, tryptophane) and derivatives thereof, imidazoles (preferably urocanic acid) and derivatives thereof, peptides, preferably D,L-carnosine, D-carnosine, L-carnosine and derivatives thereof (preferably anserine), carnitine, creatine, matrikine peptides (preferably lysyl-threonyl-threonyl-lysyl-serine) and palmitoylated pentapeptides, carotenoids, carotenes (preferably alpha-carotene, beta-carotene, lycopene) and derivatives thereof, lipoic acid and derivatives thereof (preferably dihydrolipoic acid), aurothioglucose, propyl thiouracil and other thiols (preferably thioredoxine, glutathione, cysteine, cystine, cystamine and glycosyl, N-acetyl, methyl, ethyl, propyl, amyl, butyl and lauryl, palmitoyl, oleyl, gamma-linoleyl, cholesteryl, glyceryl and oligoglyceryl esters thereof) and salts thereof, dilauryl thiodipropionate, distearyl thiodipropionate, thiodipropionic acid and derivatives thereof (preferably esters, ethers, peptides, lipids, nucleotides, nucleosides and salts) and sulfoximine compounds (preferably buthionine sulfoximines, homocysteine sulfoximine, buthionine sulfones, penta-, hexa-, heptathionine sulfoximine) in very small tolerated doses (e.g. pmol to μmol/kg), also (metal) chelators (preferably alpha-hydroxy fatty acids, palmitic acid, phytic acid, lactoferrin, alpha-hydroxy acids (preferably citric acid, lactic acid, malic acid), humic acid, bile acid, bile extracts, tannins, bilirubin, biliverdin, EDTA, EGTA and derivatives thereof), unsaturated fatty acids and derivatives thereof (preferably gamma-linolenic acid, linoleic acid, oleic acid), folic acid and derivatives thereof, ubiquinone and derivatives thereof, ubiquinol and derivatives thereof, vitamin C and derivatives (preferably ascorbyl palmitate, Mg ascorbyl phosphate, ascorbyl acetate, ascorbyl glucoside), tocopherols and derivatives (preferably vitamin E acetate), vitamin A and derivatives (vitamin A palmitate) and coniferyl benzoate of benzoic resin, rutinic acid and derivatives thereof, flavonoids and glycosylated precursors thereof, in particular quercetin and derivatives thereof, preferably alpha-glucosyl rutin, rosmarinic acid, carnosol, carnosolic acid, resveratrol, caffeic acid and derivatives thereof, sinapic acid and derivatives thereof, ferulic acid and derivatives thereof, curcuminoids, chlorogenic acid and derivatives thereof, retinoids, preferably retinyl palmitate, retinol or tretinoin, ursolic acid, levulinic acid, butyl hydroxy-

toluene, butyl hydroxyanisole, nordihydroguaiac acid, nordihydroguaiaretic acid, trihydroxybutyrophenone, uric acid and derivatives thereof, mannose and derivatives thereof, zinc and derivatives thereof (preferably ZnO, $ZnSO_4$), selenium and derivatives thereof (preferably selenium methionine), superoxide dismutase, stilbenes and derivatives thereof (preferably stilbene oxide, trans-stilbene oxide) and the derivatives (salts, esters, ethers, sugars, nucleotides, nucleosides, peptides and lipids) of these cited active ingredients which are suitable according to the invention or extracts or fractions of plants having an antioxidant effect, preferably green tea, rooibos, honeybush, grape, rosemary, sage, melissa, thyme, lavender, olive, oats, cocoa, ginkgo, ginseng, liquorice, honeysuckle, sophora, pueraria, pinus, citrus, Phyllanthus emblica or St. John's wort, grape seeds, wheat germ, Phyllanthus emblica, coenzymes, preferably coenzyme Q10, plastoquinone and menaquinone. Preferred antioxidants are selected from the group consisting of vitamin A and derivatives, vitamin C and derivatives, tocopherol and derivatives, preferably tocopheryl acetate, and ubiquinone.

[0054] If vitamin E and/or derivatives thereof are used as the antioxidant(s), it is advantageous to choose their concentrations from the range from about 0.001 to about 10 % b.w. based on the total weight of the formulation. If vitamin A or vitamin A derivatives or carotenes or derivatives thereof are used as the antioxidant(s), it is advantageous to choose their concentrations from the range from about 0.001 to aout 10 % b.w. based on the total weight of the formulation.

[0055] **Matrix-Metalloproteinase inhibitors (MMPI).** Preferred compositions comprise matrix-metalloproteinase inhibitors, especially those inhibiting matrix-metalloproteinases enzymatically cleaving collagen, selected from the group consisting of: ursolic acid, retinyl palmitate, propyl gallate, precocenes, 6-hydroxy-7-methoxy-2,2-dimethyl-1(2H)-benzopyran, 3,4-dihydro-6-hydroxy-7-methoxy-2,2-dimethyl-1(2H)-benzopyran, benzamidine hydrochloride, the cysteine proteinase inhibitors N-ethylmalemide and epsilon-amino-n-caproic acid of the serinprotease inhibitors: phenylmethylsufonylfluoride, collhibin (company Pentapharm; INCI: hydrolysed rice protein), oenotherol (company Soliance; INCI: propylene glycol, aqua, Oenothera biennis root extract, ellagic acid and ellagitannins, for example from pomegranate), phosphoramidone hinokitiol, EDTA, galardin, EquiStat (company Collaborative Group; apple fruit extract, soya seed extract, ursolic acid, soya isoflavones and soya proteins), sage extracts, MDI (company Atrium; INCI: glycosaminoglycans), fermiskin (company Silab/Mawi; INCI: water and lentinus edodes extract), actimp 1.9.3 (company Expanscience/Rahn; INCI: hydrolysed lupine protein), lipobelle soyaglycone (company Mibelle; INCI: alcohol, polysorbate 80, lecithin and soy isoflavones), extracts from green and black tea and further plant extracts, which are listed in WO 02 069992 A1 (see tables 1-12 there, incorporated herein by reference), proteins or glycoproteins from soya, hydrolysed proteins from rice, pea or lupine, plant extracts which inhibit MMPs, preferably extracts from shitake mushrooms, extracts from the leaves of the Rosaceae family, sub-family Rosoideae, quite particularly extracts of blackberry leaf (preferably as described in WO 2005 123101 A1, incorporated herein by reference) as e.g. SymMatrix (company Symrise, INCI: Maltodextrin, Rubus Fruticosus (Blackberry) Leaf Extract). Preferred actives of are selected from the group consisting of retinyl palmitate, ursolic acid, extracts from the leaves of the Rosaceae family, sub-family Rosoideae, genistein and daidzein.

[0056] **Skin-moisturizing agents.** Preferred skin moisturizing agents are selected from the group consisting of alkane diols or alkane triols comprising 3 to 12 carbon atoms, preferably C_3-C_{10}-alkane diols and C_3-C_{10}-alkane triols. More preferably the skin moisturizing agents are selected from the group consisting of: glycerol, 1,2-propylene glycol, 1,2-butylene glycol, 1,3-butylene glycol, 1,2-pentanediol, 1,2-hexanediol, 1,2-octanediol and 1,2-decanediol.

[0057] **Glycosaminoglycan stimulators.** Preferred compositions comprise substances stimulating the synthesis of glycosaminoglycans selected from the group consisting of hyaluronic acid and derivatives or salts, Subliskin (Sederma, INCI: Sinorhizobium Meliloti Ferment Filtrate, Cetyl Hydroxyethylcellulose, Lecithin), Hyalufix (BASF, INCI: Water, Butylene Glycol, Alpinia galanga leaf extract, Xanthan Gum, Caprylic/Capric Triglyceride), Stimulhyal (Soliance, INCI: Calcium ketogluconate), Syn-Glycan (DSM, INCI: Tetradecyl Aminobutyroylvalylaminobutyric Urea Trifluoroacetate, Glycerin, Magnesium chloride), Kalpariane (Biotech Marine), DC Upregulex (Distinctive Cosmetic Ingredients, INCI: Water, Butylene Glycol, Phospholipids, Hydrolyzed Sericin), glucosamine, N-acetyl glucosamine, retinoids, preferably retinol and vitamin A, Arctium lappa fruit extract, Eriobotrya japonica extract, Genkwanin, N-Methyl-L-serine, (-)-alpha-bisabolol or synthetic alpha-bisabolol such as e.g. Dragosantol and Dragosantol 100 from Symrise, oat glucan, Echinacea purpurea extract and soy protein hydrolysate. Preferred actives are selected from the group consisting of hyaluronic acid and derivatives or salts, retinol and derivatives, (-)-alpha-bisabolol or synthetic alpha-bisabolol such as e.g. Dragosantol and Dragosantol 100 from Symrise, oat glucan, Echinacea purpurea extract, Sinorhizobium Meliloti Ferment Filtrate, Calcium ketogluconate, Alpinia galanga leaf extract and tetradecyl aminobutyroylvalylaminobutyric urea trifluoroacetate.

[0058] **Anti-inflammatory agents.** The compositions may also contain anti-inflammatory and/or redness and/or itch ameliorating ingredients, in particular steroidal substances of the corticosteroid type selected from the group consisting of hydrocortisone, dexamethasone, dexamethasone phosphate, methyl prednisolone or cortisone, are advantageously used as anti-inflammatory active ingredients or active ingredients to relieve reddening and itching, the list of which can be extended by the addition of other steroidal anti-inflammatories. Non-steroidal anti-inflammatories can also be used. Examples which can be cited here are oxicams such as piroxicam or tenoxicam; salicylates such as aspirin, disalcid, solprin or fendosal; acetic acid derivatives such as diclofenac, fenclofenac, indomethacin, sulindac, tolmetin or clindanac; fenamates such as mefenamic, meclofenamic, flufenamic or niflumic; propionic acid derivatives such as ibuprofen,

naproxen, benoxaprofen or pyrazoles such as phenylbutazone, oxyphenylbutazone, febrazone or azapropazone. Anthranilic acid derivatives, in particular avenanthramides described in WO 2004 047833 A1, are preferred anti-itch ingredients in a composition according to the present invention.

[0059] Also useful are natural or naturally occurring anti-inflammatory mixtures of substances or mixtures of substances that alleviate reddening and/or itching, in particular extracts or fractions from camomile, Aloe vera, Commiphora species, Rubia species, willow, willow-herb, oats, calendula, arnica, St John's wort, honeysuckle, rosemary, Passiflora incarnata, witch hazel, ginger or Echinacea; preferably selected from the group consisting of extracts or fractions from camomile, Aloe vera, oats, calendula, arnica, honeysuckle, rosemary, witch hazel, ginger or Echinacea, and/or pure substances, preferably alpha-bisabolol, apigenin, apigenin-7-glucoside, gingerols, shogaols, gingerdiols, dehydrogingerdiones, paradols, natural or naturally occuring avenanthramides, preferably tranilast, avenanthramide A, avenanthramide B, avenanthramide C, non-natural or non-naturally occuring avenanthramides, preferably dihydroavenanthramide D, dihydroavenanthramide E, avenanthramide D, avenanthramide E, avenanthramide F, boswellic acid, phytosterols, glycyrrhizin, glabridin and licochalcone A; preferably selected from the group consisting of alpha-bisabolol, natural avenanthramides, non-natural avenanthramides, preferably dihydroavenanthramide D (as described in WO 2004 047833 A1), boswellic acid, phytosterols, glycyrrhizin, and licochalcone A, and/or allantoin, panthenol, lanolin, (pseudo-)ceramides [preferably Ceramide 2, hydroxypropyl bispalmitamide MEA, cetyloxypropyl glyceryl methoxypropyl myristamide, N-(1-hexadecanoyl)-4-hydroxy-L-proline (1-hexadecyl) ester, hydroxyethyl palmityl oxyhydroxypropyl palmitamide], glycosphingolipids, phytosterols, chitosan, mannose, lactose and β-glucans, in particular 1,3-1,4-β-glucan from oats.

[0060] When bisabolol is used in the context of the present invention it can be of natural or synthetic origin, and is preferably "alpha-bisabolol". Preferably, the bisabolol used is synthetically prepared or natural (-)-alpha-bisabolol and/or synthetic mixed-isomer alpha-bisabolol. If natural (-)-alpha-bisabolol is used, this can also be employed as a constituent of an essential oil or of a plant extract or of a fraction thereof, for example as a constituent of (fractions of) oil or extracts of camomile or of Vanillosmopsis (in particular Vanillosmopsis erythropappa or Vanillosmopsis arborea). Synthetic alpha-bisabolol is obtainable, for example, under the name "Dragosantol" from Symrise.

[0061] In case ginger extract is used in the context of the present invention, preferably extracts of the fresh or dried ginger root are used which are prepared by extraction with methanol, ethanol, iso-propanol, acetone, ethyl acetate, carbon dioxide (CO2), hexane, methylene chloride, chloroform or other solvents or solvent mixtures of comparable polarity. The extracts are characterized by the presence of active skin irritation-reducing amounts of constituents such as e.g. gingerols, shogaols, gingerdiols, dehydrogingerdiones and/or paradols.

[0062] **TRPV1 antagonists.** Suitable compounds which reduce the hypersensitivity of skin nerves based on their action as TRPV1 antagonists, encompass e.g. trans-4-tert-butyl cyclohexanol as described in WO 2009 087242 A1, or indirect modulators of TRPV1 by an activation of the μ-receptor, e.g. acetyl tetrapeptide-15, are preferred.

[0063] **Desquamating agents.** The compositions may also contain desquamating agents (component b5) in amounts of about 0.1 to about 30 % b.w. preferably about 0.5 to about 15 % b.w., particularly preferably about 1 to about 10 % b.w. based on the total weight of the preparation. The expression "desquamating agent" is understood to mean any compound capable of acting:

- either directly on desquamation by promoting exfoliation, such as β-hydroxy acids, in particular salicylic acid and its derivatives (including 5-n-octanoylsalicylic acid); α-hydroxy acids, such as glycolic, citric, lactic, tartaric, malic or mandelic acids; urea; gentisic acid; oligofucoses; cinnamic acid; extract of Sophora japonica; resveratrol and some derivatives of jasmonic acid;

- or on the enzymes involved in the desquamation or the degradation of the corneodesmosomes, glycosidases, stratum corneum chymotryptic enzyme (SCCE) or other proteases (trypsin, chymotrypsin-like). There may be mentioned agents chelating inorganic salts: EDTA; N-acyl-N,N',N'-ethylenediaminetriacetic acid; aminosulphonic compounds and in particular (N-2-hydroxyethylpiperazine-N-2-ethane)sulphonic acid (HEPES); derivatives of 2-oxothiazolidine-4-carboxylic acid (procysteine); derivatives of alpha-amino acids of the glycine type (as described in EP-0 852 949, and sodium methylglycine diacetate marketed by BASF under the trade name TRILON M); honey; sugar derivatives such as O-octanoyl-6-D-maltose and N-acetylglucosamine; chestnut extracts such as those marketed by the company SILAB under the name Recoverine®, prickly pear extracts such as those marketed under the name Exfolactive® by the company SILAB, or Phytosphingosine SLC® (phytosphingosine grafted with a salicylic acid) marketed by the company Degussa.

[0064] Desquamating agents suitable for the invention may be chosen in particular from the group comprising sulphonic acids, calcium chelators, α-hydroxy acids such as glycolic, citric, lactic, tartaric, malic or mandelic acids; ascorbic acid and its derivatives such as ascorbyl glucoside and magnesium ascorbyl phosphate; nicotinamide; urea; (N-2-hydroxyethylpiperazine-N-2-ethane)sulphonic acid (HEPES), β-hydroxy acids such as salicylic acid and its derivatives, retinoids such as retinol and its esters, retinal, retinoic acid and its derivatives, those described in the **documents** FR 2570377

A1, EP 0199636 A1, EP 0325540 A1, EP 0402072 A1, chestnut or prickly pear extracts, in particular marketed by SILAB; reducing compounds such as cysteine or cysteine precursors.

[0065] Desquamating agents which can be used are also nicotinic acid and its esters and nicotinamide, also called vitamin B3 or vitamin PP, and ascorbic acid and its precursors, as described in particular in application EP 1529522 A1.

[0066] **Anti-cellulite agents.** Anti-cellulite agents and lipolytic agents are preferably selected from the group consisting of those described in WO 2007/077541, and beta-adrenergic receptor agonists such as synephrine and its derivatives, and cyclohexyl carbamates described in WO 2010/097479. Agents enhancing or boosting the activity of anti-cellulite agents, in particular agents which stimulate and/or depolarise C nerve fibres, are preferably selected from the group consisting of capsaicin and derivatives thereof, vanillyl-nonylamid and derivatives thereof, L-carnitine, coenzym A, isoflavonoides, soy extracts, ananas extract and conjugated linoleic acid.

[0067] **Fat enhancing agents.** Formulations and products according to the present invention may also comprise one or more fat enhancing and/or adipogenic agents as well as agents enhancing or boosting the activity of fat enhancing agents. A fat enhancing agent is for example hydroxymethoxyphenyl propylmethylmethoxybenzofuran (trade name: Sym3D®).

HAIR GROWTH ACTIVATORS OR INHIBITORS

[0068] Formulations and products according to the present invention may also comprise one or more hair growth activators, i.e. agents to stimulate hair growth. Hair growth activators are preferably selected from the group consisting of pyrimidine derivatives such as 2,4-diaminopyrimidine-3-oxide (Aminexil), 2,4-diamino-6-piperidinopyrimidine-3-oxide (Minoxidil) and derivatives thereof, 6-amino-1,2-dihydro-1-hydroxy-2-imino-4-piperidinopyrimidine and its derivatives, xanthine alkaloids such as caffeine, theobromine and theophylline and derivatives thereof, quercetin and derivatives, dihydroquercetin (taxifolin) and derivatives, potassium channel openers, antiandrogenic agents, synthetic or natural 5-reductase inhibitors, nicotinic acid esters such as tocopheryl nicotinate, benzyl nicotinate and C1-C6 alkyl nicotinate, proteins such as for example the tripeptide Lys-Pro-Val, diphencypren, hormons, finasteride, dutasteride, flutamide, bicalutamide, pregnane derivatives, progesterone and its derivatives, cyproterone acetate, spironolactone and other diuretics, calcineurin inhibitors such as FK506 (Tacrolimus, Fujimycin) and its derivatives, Cyclosporin A and derivatives thereof, zinc and zinc salts, polyphenols, procyanidins, proanthocyanidins, phytosterols such as for example beta-sitosterol, biotin, eugenol, (±)-beta-citronellol, panthenol, glycogen for example from mussels, extracts from microorganisms, algae, plants and plant parts of for example the genera dandelion (Leontodon or Taraxacum), Orthosiphon, Vitex, Coffea, Paullinia, Theobroma, Asiasarum, Cucurbita or Styphnolobium, Serenoa repens (saw palmetto), Sophora flavescens, Pygeum africanum, Panicum miliaceum, Cimicifuga racemosa, Glycine max, Eugenia caryophyllata, Cotinus coggygria, Hibiscus rosa-sinensis, Camellia sinensis, Ilex paraguariensis, Isochrysis galbana, licorice, grape, apple, barley or hops or/nd hydrolysates from rice or wheat.

[0069] Alternatively, formulations and products according to the present invention may comprise one or more hair growth inhibitors (as described above), i.e. agents to reduce or prevent hair growth. Hair growth inhibitors are preferably selected from the group consisting of activin, activin derivatives or activin agonists, ornithine decarboxylase inhibitors such as alpha-difluoromethylornithine or pentacyclic triterpenes like for example ursolic acid, betulin, betulinic acid, oleanolic acid and derivatives thereof, 5alpha-reductase inhibitors, androgen receptor antagonists, S-adenosylmethionine decarboxylase inhibitors, gamma-glutamyl transpeptidase inhibitors, transglutaminase inhibitors, soybean-derived serine protease inhibitors, extracts from microorganisms, algae, different microalgae or plants and plant parts of for example the families Leguminosae, Solanaceae, Graminae, Asclepiadaceae or Cucurbitaceae, the genera Chondrus, Gloiopeltis, Ceramium, Durvillea, Glycine max, San-guisorba officinalis, Calendula officinalis, Hamamelis virginiana, Arnica montana, Salix alba, Hypericum perforatum or Gymnema sylvestre.

COOLING AGENTS

[0070] The compositions may also contain one or more substances with a physiological cooling effect (cooling agents), which are preferably selected here from the following list: menthol and menthol derivatives (for example L-menthol, D-menthol, racemic menthol, isomenthol, neoisomenthol, neomenthol) menthylethers (for example (l-menthoxy)-1,2-propandiol, (l-menthoxy)-2-methyl-1,2-propandiol, l-menthyl-methylether), menthylesters (for example menthylformiate, menthylacetate, menthylisobutyrate, menthyllactates, L-menthyl-L-lactate, L-menthyl-D-lactate, menthyl-(2-methoxy)acetate, menthyl-(2-methoxyethoxy)acetate, menthylpyroglutamate), menthylcarbonates (for example menthylpropyleneglycolcarbonate, menthylethyleneglycolcarbonate, menthylglycerolcarbonate or mixtures thereof), the semi-esters of menthols with a dicarboxylic acid or derivatives thereof (for example mono-menthylsuccinate, mono-menthylglutarate, mono-menthylmalonate, O-menthyl succinic acid ester-N,N-(dimethyl)amide, O-menthyl succinic acid ester amide), menthanecarboxylic acid amides (in this case preferably menthanecarboxylic acid-N-ethylamide [WS3] or N^{α}-(menthanecarbonyl)glycinethylester [WS5], as described in US 4,150,052, menthanecarboxylic acid-N-(4-cyanophe-

nyl)amide or menthanecarboxylic acid-N-(4-cyanomethylphenyl)amide as described in WO 2005 049553 A1, methanecarboxylic acid-N-(alkoxyalkyl)amides), menthone and menthone derivatives (for example L-menthone glycerol ketal), 2,3-dimethyl-2-(2-propyl)-butyric acid derivatives (for example 2,3-dimethyl-2-(2-propyl)-butyric acid-N-methylamide [WS23]), isopulegol or its esters (l-(-)-isopulegol, l-(-)-isopulegolacetate), menthane derivatives (for example p-menthane-3,8-diol), cubebol or synthetic or natural mixtures, containing cubebol, pyrrolidone derivatives of cycloalkyldione derivatives (for example 3-methyl-2(1-pyrrolidinyl)-2-cyclopentene-1-one) or tetrahydropyrimidine-2-one (for example iciline or related compounds, as described in WO 2004/026840), further carboxamides (for example N-(2-(pyridin-2-yl)ethyl)-3-p-menthanecarboxamide or related compounds), (1R,2S,5R)-N-(4-Methoxyphenyl)-5-methyl-2-(1-isopropyl)cyclohexane-carboxamide [WS12], oxamates (preferably those described in EP 2033688 A2).

ANTI-MICROBIAL AGENTS

[0071] Suitable anti-microbial agents are, for example, 4- hydroxybenzoic acid and its salts and esters, 4-hydroxy acetophenone, 4-methylbenzyl alcohol, troplone, hinokitiol, N-(4-chlorophenyl)-N'-(3,4- dichlorophenyl)urea, 2,4,4'-trichloro-2'-hydroxy-diphenyl ether (triclosan), 4-chloro-3,5-dimethyl-phenol, 2,2'-methylenebis(6-bromo-4- chlorophenol), 3-methyl-4-(1-methylethyl)phenol, 2-benzyl-4-chloro-phenol, 3-(4-chlorophenoxy)-1,2-propanediol, 3-iodo-2-propynyl butylcarbamate, chlorhexidine, 3,4,4'-trichlorocarbanilide (TTC), antibacterial fragrances, thymol, thyme oil, eugenol, oil of cloves, menthol, mint oil, farnesol, phenoxyethanol, glycerol monocaprate, glycerol monocaprylate, glycerol monolaurate (GML), diglycerol monocaprate (DMC), salicylic acid N-alkylamides, such as, for example, n-octylsalicylamide or n- decylsalicylamide. Further antimicrobials are chitosan, totarol, arylalkyl alcohols, such as e.g. 4-methyl-4-phenyl-2-pentanol and its derivatives (DE 101 43 434, in particular 4-methyl-4-phenyl-2-pentanol), muguet alcohol (2,2-dimethyl-3-phenylpropanol), other arylalkyl alcohols (e.g. as disclosed in DE 44 47 361, DE 103 30 697, US 4,110,430 or EP 1157 687), 2-butyloctanoic acid, 2-hexyldecanoic acid, p-anisic acid, perfume oils or single aroma chemicals with antimicrobial activity, polyglycerol esters, such as e.g. polyglyceryl 3-caprylates, or combinations of the substances mentioned, which are generally employed, inter alia, against underarm odour, foot odour, acne or dandruff formation.

ENZYME INHIBITORS

[0072] Suitable enzyme inhibitors are, for example, esterase inhibitors. These are preferably trialkyl citrates, such as trimethyl citrate, tripropyl citrate, triisopropyl citrate, tributyl citrate and, in particular, triethyl citrate (Hydagen CAT). The substances inhibit enzyme activity, thereby reducing the formation of odour. Other substances which are suitable esterase inhibitors are sterol sulfates or phosphates, such as, for example, lanosterol, cholesterol, campesterol, stigmasterol and sitosterol sulfate or phosphate, dicarboxylic acids and esters thereof, such as, for example, glutaric acid, monoethyl glutarate, diethyl glutarate, adipic acid, monoethyl adipate, diethyl adipate, malonic acid and diethyl malonate, hydroxycarboxylic acids and esters thereof, such as, for example, citric acid, malic acid, tartaric acid or diethyl tartrate, and zinc glycinate.

ODOUR ABSORBERS AND ANTIPERSPIRANT ACTIVE AGENTS

[0073] Suitable odour absorbers are substances which are able to absorb and largely retain odour-forming compounds. They lower the partial pressure of the individual components, thus also reducing their rate of diffusion. It is important that perfumes must remain unimpaired in this process. Odour absorbers are not effective against bacteria. They comprise, for example, as main constituent, a complex zinc salt of ricinoleic acid or specific, largely odour-neutral fragrances which are known to the person skilled in the art as "fixatives", such as, for example, extracts of labdanum or styrax or certain abietic acid derivatives. The odour masking agents are fragrances or perfume oils, which, in addition to their function as odour masking agents, give the deodorants their respective fragrance note. Perfume oils which may be mentioned are, for example, mixtures of natural and synthetic fragrances. Natural fragrances are extracts from flowers, stems and leaves, fruits, fruit peels, roots, woods, herbs and grasses, needles and branches, and resins and balsams. Also suitable are animal products, such as, for example, civet and castoreum. Typical synthetic fragrance compounds are products of the ester, ether, aldehyde, ketone, alcohol, and hydrocarbon type. Fragrance compounds of the ester type are, for example, benzyl acetate, p-tert-butylcyclohexyl acetate, linalyl acetate, phenylethyl acetate, linalyl benzoate, benzyl formate, allyl cyclohexylpropionate, styrallyl propionate and benzyl salicylate. The ethers include, for example, benzyl ethyl ether, and the aldehydes include, for example, the linear alkanals having 8 to 18 carbon atoms, citral, citronellal, citronellyloxyacetaldehyde, cyclamen aldehyde, hydroxycitronellal, lilial and bourgeonal, the ketones include, for example, the ionones and methyl cedryl ketone, the alcohols include anethole, citronellol, eugenol, isoeugenol, geraniol, linaool, phenylethyl alcohol and terpineol, and the hydrocarbons include mainly the terpenes and balsams. Preference is, however, given to using mixtures of different fragrances which together produce a pleasing fragrance note. Essential oils of relatively low volatility, which are mostly used as aroma components, are also suitable as perfume oils, e.g. sage

oil, camomile oil, oil of cloves, melissa oil, mint oil, cinnamon leaf oil, linden flower oil, juniperberry oil, vetiver oil, olibanum oil, galbanum oil, labdanum oil and lavandin oil. Preference is given to using bergamot oil, dihydromyrcenol, lilial, lyral, citronellol, phenylethyl alcohol, α-hexylcinnamaldehyde, geraniol, benzylacetone, cyclamen aldehyde, linalool, boisambrene forte, ambroxan, indole, hedione, sandelice, lemon oil, mandarin oil, orange oil, allyl amyl glycolate, cyclovertal, lavandin oil, clary sage oil, β- damascone, geranium oil bourbon, cyclohexyl salicylate, Vertofix coeur, iso-E-super, Fixolide NP, evernyl, iraldein gamma, phenylacetic acid, geranyl acetate, benzyl acetate, rose oxide, romilat, irotyl and floramat alone or in mixtures.

[0074] Suitable astringent antiperspirant active ingredients are primarily salts of aluminium, zirconium or of zinc. Such suitable antihydrotic active ingredients are, for example, aluminium chloride, aluminium chlorohydrate, aluminium dichlorohydrate, aluminium sesquichlorohydrate and complex compounds thereof, e.g. with 1,2- propylene glycol, aluminium hydroxyallantoinate, aluminium chloride tartrate, aluminium zirconium trichlorohydrate, aluminium zirconium tetrachlorohydrate, aluminium zirconium pentachlorohydrate and complex compounds thereof, e.g. with amino acids, such as glycine.

FILM FORMERS AND ANTI-DANDRUFF AGENTS

[0075] Standard film formers are, for example, chitosan, microcrystalline chitosan, quaternized chitosan, polyvinyl pyrrolidone, vinyl pyrrolidone/vinyl acetate copolymers, polymers of the acrylic acid series, quaternary cellulose derivatives, collagen, hyaluronic acid and salts thereof and similar compounds.

[0076] Suitable antidandruff agents are Pirocton Olamin (1-hydroxy-4-methyl-6-(2,4,4-trimethylpentyl)-2-(1H)-pyridinone monoethanolamine salt), Baypival (Climbazole), Ketoconazol® (4-acetyl-1-{4-[2-(2,4-dichlorophenyl) r-2-(1H-imidazol-1-ylmethyl)-1,3-dioxylan-c-4-ylmethoxyphenyl}-piperazine, ketoconazole, elubiol, selenium disulfide, colloidal sulfur, sulfur polyethylene glycol sorbitan monooleate, sulfur ricinol polyethoxylate, sulfur tar distillate, salicylic acid (or in combination with hexachlorophene), undecylenic acid, monoethanolamide sulfosuccinate Na salt, Lamepon® UD (protein/undecylenic acid condensate), zinc pyrithione, aluminium pyrithione and magnesium pyrithione/dipyrithione magnesium sulfate.

CARRIERS AND HYDROTROPES

[0077] Preferred cosmetics carrier materials are solid or liquid at 25°C and 1013 mbar (including highly viscous substances) as for example glycerol, 1,2-propylene glycol, 1,2-butylene glycol, 1,3-propylene glycol, 1,3-butylene glycol, ethanol, water and mixtures of two or more of said liquid carrier materials with water. Optionally, these preparations according to the invention may be produced using preservatives or solubilizers. Other preferred liquid carrier substances, which may be a component of a preparation according to the invention are selected from the group consisting of oils such as vegetable oil, neutral oil and mineral oil.

[0078] Preferred solid carrier materials, which may be a component of a preparation according to the invention are hydrocolloids, such as starches, degraded starches, chemically or physically modified starches, dextrins, (powdery) maltodextrins (preferably with a dextrose equivalent value of 5 to 25, preferably of 10 - 20), lactose, silicon dioxide, glucose, modified celluloses, gum arabic, ghatti gum, traganth, karaya, carrageenan, pullulan, curdlan, xanthan gum, gellan gum, guar flour, carob bean flour, alginates, agar, pectin and inulin and mixtures of two or more of these solids, in particular maltodextrins (preferably with a dextrose equivalent value of 15 - 20), lactose, silicon dioxide and/or glucose.

[0079] In addition, hydrotropes, for example ethanol, isopropyl alcohol or polyols, may be used to improve flow behaviour. Suitable polyols preferably contain 2 to 15 carbon atoms and at least two hydroxyl groups. The polyols may contain other functional groups, more especially amino groups, or may be modified with nitrogen. Typical examples are

- glycerol;
- alkylene glycols such as, for example, ethylene glycol, diethylene glycol, propylene glycol, butylene glycol, hexylene glycol and polyethylene glycols with an average molecular weight of 100 to 1000 Dalton;
- technical oligoglycerol mixtures with a degree of self-condensation of 1.5 to 10, such as for example technical diglycerol mixtures with a diglycerol content of 40 to 50% by weight;
- methylol compounds such as, in particular, trimethylol ethane, trimethylol propane, trimethylol butane, pentaerythritol and dipentaerythritol;
- lower alkyl glucosides, particularly those containing 1 to 8 carbon atoms in the alkyl group, for example methyl and butyl glucoside;

- sugar alcohols containing 5 to 12 carbon atoms, for example sorbitol or mannitol,
- sugars containing 5 to 12 carbon atoms, for example glucose or sucrose;
- amino sugars, for example glucamine;
- dialcoholamines, such as diethanolamine or 2-aminopropane-1,3-diol.

PRESERVATIVES

[0080] Suitable preservatives are, for example, phenoxyethanol, formaldehyde solution, parabens, pentanediol or sorbic acid and the other classes of compounds listed in Appendix 6, Parts A and B of the Kosmetikverordnung ("Cosmetics Directive").

PERFUME OILS AND FRAGRANCES

[0081] Suitable perfume oils are mixtures of natural and synthetic perfumes. Natural perfumes include the extracts of blossoms (lily, lavender, rose, jasmine, neroli, ylang-ylang), stems and leaves (geranium, patchouli, petitgrain), fruits (anise, coriander, caraway, juniper), fruit peel (bergamot, lemon, orange), roots (nutmeg, angelica, celery, cardamom, costus, iris, calmus), woods (pinewood, sandalwood, guaiac wood, cedarwood, rosewood), herbs and grasses (tarragon, lemon grass, sage, thyme), needles and branches (spruce, fir, pine, dwarf pine), resins and balsams (galbanum, elemi, benzoin, myrrh, olibanum, opoponax). Animal raw materials, for example civet and beaver, may also be used. Typical synthetic perfume compounds are products of the ester, ether, aldehyde, ketone, alcohol and hydrocarbon type. Examples of perfume compounds of the ester type are benzyl acetate, phenoxyethyl isobutyrate, p-tert.butyl cyclohexylacetate, linalyl acetate, dimethyl benzyl carbinyl acetate, phenyl ethyl acetate, linalyl benzoate, benzyl formate, ethylmethyl phenyl glycinate, allyl cyclohexyl propionate, styrallyl propionate and benzyl salicylate. Ethers include, for example, benzyl ethyl ether while aldehydes include, for example, the linear alkanals containing 8 to 18 carbon atoms, citral, citronellal, citronellyloxyacetaldehyde, cyclamen aldehyde, hydroxycitronellal, lilial and bourgeonal. Examples of suitable ketones are the ionones, ▪ -isomethylionone and methyl cedryl ketone. Suitable alcohols are anethol, citronellol, eugenol, isoeugenol, geraniol, linalool, phenylethyl alcohol and terpineol. The hydrocarbons mainly include the terpenes and balsams. However, it is preferred to use mixtures of different perfume compounds which, together, produce an agreeable perfume. Other suitable perfume oils are essential oils of relatively low volatility which are mostly used as aroma components. Examples are sage oil, camomile oil, clove oil, melissa oil, mint oil, cinnamon leaf oil, lime-blossom oil, juniper berry oil, vetiver oil, olibanum oil, galbanum oil, ladanum oil and lavendin oil. The following are preferably used either individually or in the form of mixtures: bergamot oil, dihydromyrcenol, lilial, lyral, citronellol, phenylethyl alcohol, hexylcinnamaldehyde, geraniol, benzyl acetone, cyclamen aldehyde, linalool, Boisambrene Forte, Ambroxan, indole, hedione, sandelice, citrus oil, mandarin oil, orange oil, allylamyl glycolate, cyclovertal, lavendin oil, clary oil, damascone, geranium oil bourbon, cyclohexyl salicylate, Vertofix Coeur, Iso-E-Super, Fixolide NP, evernyl, iraldein gamma, phenylacetic acid, geranyl acetate, benzyl acetate, rose oxide, romillat, irotyl and floramat.

DYES

[0082] Suitable dyes are any of the substances suitable and approved for cosmetic purposes as listed, for example, in the publication **"Kosmetische Färbemittel"** of the Farbstoffkommission der Deutschen Forschungsgemeinschaft, **Verlag Chemie, Weinheim, 1984, pages 81 to 106.** Examples include cochineal red A (C.I. 16255), patent blue V (C.I. 42051), indigotin (C.I. 73015), chlorophyllin (C.I. 75810), quinoline yellow (C.I. 47005), titanium dioxide (C.I. 77891), indanthrene blue RS (C.I. 69800) and madder lake (C.I. 58000). Luminol may also be present as a luminescent dye. Advantageous coloured pigments are for example titanium dioxide, mica, iron oxides (e.g. Fe_2O_3 Fe_3O_4, FeO(OH)) and/or tin oxide. Advantageous dyes are for example carmine, Berlin blue, chromium oxide green, ultramarine blue and/or manganese violet.

PREPARATIONS

[0083] Preferred compositions according to the present inventions are selected from the group of products for treatment, protecting, care and cleansing of the skin and/or hair or as a make-up product, preferably as a leave-on product (meaning that the one or more compounds of formula (I) stay on the skin and/or hair for a longer period of time, compared to rinse-off products, so that the moisturizing and/or anti-ageing and/or wound healing promoting action thereof is more pronounced).

[0084] The formulations according to the invention are preferably in the form of an emulsion, e.g. W/O (water-in-oil), O/W (oil-in-water), W/O/W (water-in-oil-in-water), O/W/O (oil-in-water-in-oil) emulsion, PIT emulsion, Pickering emulsion, emulsion with a low oil content, micro- or nanoemulsion, a solution, e.g. in oil (fatty oils or fatty acid esters, in particular C_6-C_{32} fatty acid C_2-C_{30} esters) or silicone oil, dispersion, suspension, creme, lotion or milk, depending on the production method and ingredients, a gel (including hydrogel, hydrodispersion gel, oleogel), spray (e.g. pump spray or spray with propellant) or a foam or an impregnating solution for cosmetic wipes, a detergent, e.g. soap, synthetic detergent, liquid washing, shower and bath preparation, bath product (capsule, oil, tablet, salt, bath salt, soap, etc.), effervescent preparation, a skin care product such as e.g. an emulsion (as described above), ointment, paste, gel (as described above), oil, balsam, serum, powder (e.g. face powder, body powder), a mask, a pencil, stick, roll-on, pump, aerosol (foaming, non-foaming or post-foaming), a deodorant and/or antiperspirant, mouthwash and mouth rinse, a foot care product (including keratolytic, deodorant), an insect repellent, a sunscreen, aftersun preparation, a shaving product, aftershave balm, pre- and aftershave lotion, a depilatory agent, a hair care product such as e.g. shampoo (including 2-in-1 shampoo, anti-dandruff shampoo, baby shampoo, shampoo for dry scalps, concentrated shampoo), conditioner, hair tonic, hair water, hair rinse, styling creme, pomade, perm and setting lotion, hair spray, styling aid (e.g. gel or wax), hair smoothing agent (detangling agent, relaxer), hair dye such as e.g. temporary direct-dyeing hair dye, semi-permanent hair dye, permanent hair dye, hair conditioner, hair mousse, eye care product, make-up, make-up remover or baby product.

[0085] The formulations according to the invention are particularly preferably in the form of an emulsion, in particular in the form of a W/O, O/W, W/O/W, O/W/O emulsion, PIT emulsion, Pickering emulsion, emulsion with a low oil content, micro- or nanoemulsion, a gel (including hydrogel, hydrodispersion gel, oleogel), a solution e.g. in oil (fatty oils or fatty acid esters, in particular C_6-C_{32} fatty acid C_2-C_{30} esters)) or silicone oil, or a spray (e.g. pump spray or spray with propellant).

[0086] Auxiliary substances and additives can be included in quantities of 5 to 99 % b.w., preferably 10 to 80 % b.w., based on the total weight of the formulation. The amounts of cosmetic or dermatological auxiliary agents and additives and perfume to be used in each case can easily be determined by the person skilled in the art by simple trial and error, depending on the nature of the particular product.

[0087] The preparations can also contain water in a quantity of up to 99 % b.w., preferably 5 to 80 % b.w., based on the total weight of the preparation.

PHARMACEUTICAL COMPOSITIONS

[0088] Pharmaceutical compositions according to the present invention may include similar additives as already explained for the cosmetic application, such as for example oil bodies or emulsifiers and in particular co-actives supporting the beneficial properties of the new ginger extracts. It should also be mentioned that several actives cited in the following can also be incorporated in cosmetic formulations, so-called "cosmeceuticals". Therefore, the border between cosmetic and pharmaceutical compositions is in flow and it should be understood that components cited for one application are recommended for the other *mutatis-mutandis* without literal repetition.

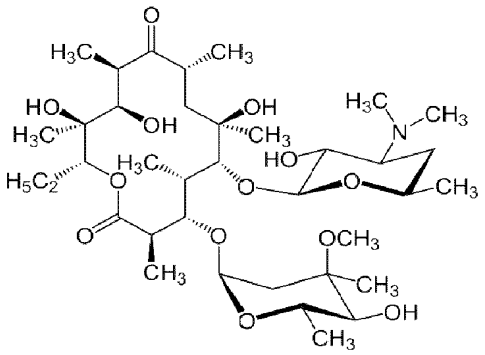
Anti-irritation agents

[0089] An important group of co-actives encompass anti-irritant agents such as for example steroidal anti-inflammatory substances of the corticosteroid type, such as e.g. hydrocortisone, hydrocortisone derivatives, such as hydrocortisone 17-butyrate, dexamethasone, dexamethasone phosphate, methylprednisolone or cortisone; non-steroidal anti-inflammatories like oxicams, such as piroxicam or tenoxicam; salicylates, such as aspirin, Disalcid, Solprin or fendosal; acetic acid derivatives, such as diclofenac, fenclofenac, indomethacin, sulindac, tolmetin or clindanac; fenamates, such as mefenamic, meclofenamic, flufenamic or niflumic; propionic acid derivatives, such as ibuprofen, naproxen or benoxaprofen, or pyrazoles, such as phenylbutazone, oxyphenylbutazone, febrazone or azapropazone. Alternatively, natural anti-inflammatory substances or reddening- and/or itching-alleviating substances can be employed. Plant extracts, specific highly active plant extract fractions and highly pure active substances isolated from plant extracts, can be employed like extracts, fractions and active substances from aloe vera, Commiphora species, Rubia species, Rubus species, willow, rose-bay, willowherb, oats, calendula, arnica, St. John's wort, honeysuckle, ginger, chamomile, rosemary, sage, melissa, Passiflora incarnata, Sophora japonica, witch hazel, Pueraria, Dianthus or Echinacea, as well as pure substances, such as, inter alia, bisabolol, apigenin, apigenin-7-glucoside, rosmarinic acid, boswellic acid, phytosterols, glycyrrhizic acid, glabridin, licochalcone A, [6]-paradol, and anthranilic acid amides, such as, in particular, avenanthramides or dianthramides, are particularly preferred. The total amount of anti-irritants in a formulation or product according to the invention is preferably in the range of from 0.0001 to 20 wt.%, preferably from 0.0001 to 10 wt.%, in particular from 0.001 to 5 wt.%, based on the total weight of the formulation or product, respectively.

[0090] Particular useful co-actives are selected from the group consisting of anti-mycotica and pain relief agents, and more particularly the group consisting of erythromycin, dimetindene, betamethasone, ibuprofen, ketoprofene, diclofenac,

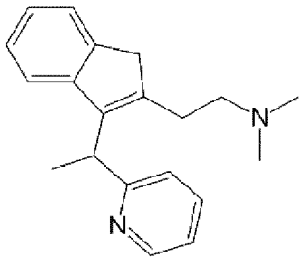
metronidazole, acyclovir, imiquimod, terbinafine, docosanol, cyclopyroxolamine, and their mixtures:

[0091] **Erythromycin** is a macrolide antibiotic that has an antimicrobial spectrum similar to or slightly wider than that of penicillin, and is often used for people who have an allergy to penicillin.

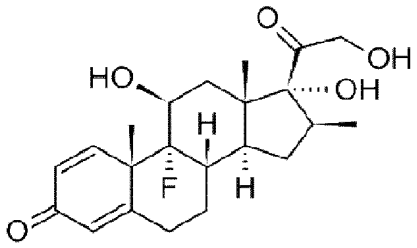


[0092] Recent studies have also shown that it can be used as a mild anti-depressant. For respiratory tract infections, it has better coverage of atypical organisms, including *Mycoplasma* and legionellosis. It was first marketed by Eli Lilly and Company, and it is today commonly known as EES (erythromycin ethylsuccinate, an ester prodrug that is commonly administered). In structure, this macrocyclic compound contains a 14-membered lactone ring with ten asymmetric centres and two sugars (L-cladinose and D-desosamine), making it a compound very difficult to produce via synthetic methods. Erythromycin is produced from a strain of the actinomycete *Saccharopolyspora erythraea* (see US 2,653,899 - Eli Lily).

[0093] **Dimetindene,** also known as Fenistil (RS-dimethyl(2-(3-[pyridin-2-yl)ethyl]-1H-inden-2-yl)ethyl)amine) is an antihistamine/anticholinergic used orally and locally as an antipruritic.

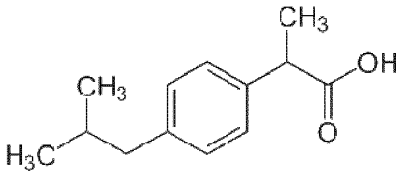


[0094] **Betamethasone** (8S,9R,10S.11S,13S,14S,16S,17R)-9-fluoro-11,17-(2-hydroxyacetyl)-10, 13,16-trimethyl-6,7,8,9,10,11,12,13,14,15,16,17-dodecahydro-3H-cyclopenta(alpha)-phenanthren-3-one) is a potent glucocorticoid steroid with anti-inflammatory and immunosuppressive properties.



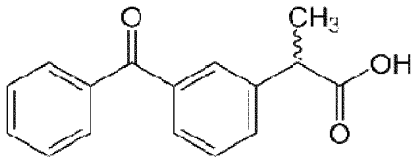
[0095] Unlike other drugs with these effects, betamethasone does not cause water retention. It is applied as a topical cream, ointment, foam, lotion or gel to treat itching. Betamethasone sodium phosphate is sometimes prescribed as an intramuscular injection (I.M) for itching from various ailments, including allergic reactions to poison ivy and similar plants (see US 3,053,865 - Merck).

[0096] **Ibuprofen** (RS)-2-(4-(2-methylpropyl)phenyl)propanoic acid) from the nomenclature iso-butyl-propanoic-phenolic acid) is a non-steroidal anti-inflammatory drug (NSAID) used for relief of symptoms of arthritis, fever, as an analgesic (pain reliever), especially where there is an inflammatory component, and dysmenorrhea.



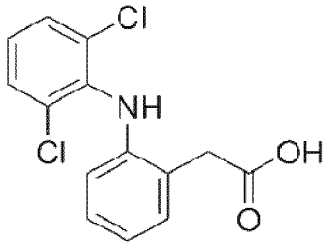
[0097] Ibuprofen is known to have an antiplatelet effect, though it is relatively mild and somewhat short-lived when compared with aspirin or other better-known antiplatelet drugs. In general, ibuprofen also acts as a vasoconstrictor, having been shown to constrict coronary arteries and some other blood vessels mainly because it inhibits the vasodilating prostacyclin produced by cyclooxygenase 2 enzymes. Ibuprofen was derived from propanoic acid by the research arm of Boots Group during the 1960s and was patented in 1961. Originally marketed as Brufen, ibuprofen is available under a variety of popular trademarks, including Motrin, Nurofen, Advil, and Nuprin (see US 3,385,886 - Boots).

[0098] **Ketoprofen** (RS)2-(3-benzoylphenyl)-propionic acid is another one of the propionic acid class of non-steroidal anti-inflammatory drug (NSAID) with analgesic and antipyretic effects.



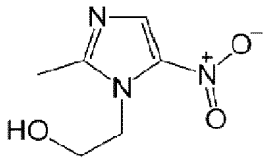
[0099] It acts by inhibiting the body's production of prostaglandins (see US 3,641,127 - Rhone-Poulenc).

[0100] **Diclofenac** is also a non-steroidal anti-inflammatory drug (NSAID) taken to reduce inflammation and as an analgesic reducing pain in certain conditions.



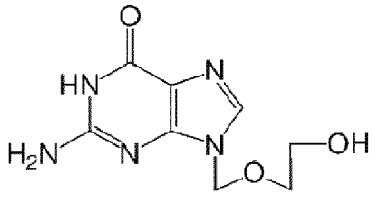
[0101] The name is derived from its chemical name: 2-(2,6-dichloranilino) phenylacetic acid. In the United Kingdom, India, Brazil and the United States, it may be supplied as either the sodium or potassium salt, in China most often as the sodium salt, while in some other countries only as the potassium salt. Diclofenac is available as a generic drug in a number of formulations. Over-the-counter (OTC) use is approved in some countries for minor aches and pains and fever associated with common infections (see US 3,558,690 - Ciba-Geigy).

[0102] **Metronidazole** (2-(2-methyl-5-nitro-1H-imidazol-1-yl)ethanol) is a nitroimidazole antibiotic medication used particularly for anaerobic bacteria and protozoa.



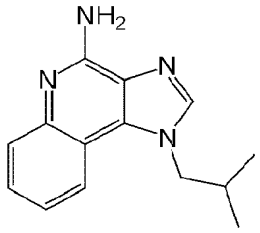
[0103] Metronidazole is an antibiotic, amebicide, and antiprotozoal. It is the drug of choice for first episodes of mild-to-moderate *Clostridium difficile* infection. It is marketed in the U.S.A. by Pfizer and globally by Sanofiunder the trade name Flagyl, in Pakistan and Bangladesh also as Nidagyl by Star Laboratories, and in Thailand, as Mepagyl by Thai Nakhorn Patana. It is also marketed in UK by Milpharm Limited and Almus Pharmaceuticals. Metronidazole was developed in 1960. Metronidazole is used also as a gel preparation in the treatment of the dermatological conditions such as rosaceae and fungating tumours (see US 2,944,061 - Rhone Poulenc).

[0104] **Acyclovir** or acyclovir (USAN, former BAN), chemical name acycloguanosine (2-Amino-1,9-dihydro-9-((2-hydroxyethoxy)methyl)-6H-Purin-6-one), abbreviated as ACV is a guanosine analogue antiviral drug, marketed under trade names such as *Cyclovir, Herpex, Acivir, Acivirax, Zovirax,* and *Xovir.* The solid active agent has a solubility in water (20° dH) at 20 °C of less than 5 g/L.



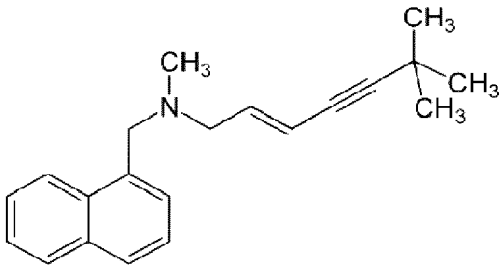
[0105] One of the most commonly used antiviral drugs; it is primarily used for the treatment of herpes simplex virus infections, as well as in the treatment of varicella zoster (chickenpox) and herpes zoster (shingles); see also US 4,199,574 (Wellcome).

[0106] **Imiquimod** (3-(2-methylpropyl)-3,5,8-triazatricyclo[7.4.0.0.^{2,6}]trideca-1(9),2(6),4,7,10, 12- hexaen-7-amine, INN) is a prescription medication that acts as an immune response modifier.



[0107] It is marketed by Meda AB, Graceway Pharmaceuticals and iNova Pharmaceuticals under the trade names Aldara and Zyclara, and by Mochida as Beselna. It is also referred to as R-837 (see US 4,689,338 - Riker).

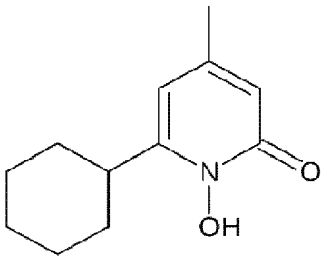
[0108] **Terbinafine,** more particularly terbinafine hydrochloride [(2E)-6,6-dimethylhept-2-en-4-yn-1-yl](methyl)(naphthalen-1-ylmethyl)amine) is a synthetic allylamine antifungal from Novartis. It is highly lipophilic in nature and tends to accumulate in skin, nails, and fatty tissues.



[0109] It is sold by the name Lamisil in Argentina, Australia, Belgium, Brazil, Canada, Chile, Egypt, Finland, France, Germany, Greece, Hungary, Iceland, Ireland, Israel, Mexico, Pakistan, Peru, New Zealand, Norway, Romania, Russia, Slovenia, South Africa, Sweden, United Kingdom, United States and Venezuela, also sold under the name Corbinal and Terbisil in Turkey and under the name "undofen cream" in Poland. As a generic it is sold under the name Zabel in Australia. It is also available as a generic medication in the United States, United Kingdom, Belgium, Switzerland and Brazil. In India, Terbinafine hydrochloride is available in topical form under the brand name Sebifin (Ranbaxy Labs), Zimig (GSK Pharma) and mycoCeaze (Progres Laboratories).MycoVa, developed by Apricus Biosciences, is a topical nail solution of terbinafine and DDAIP which has completed three Phase III studies for the treatment of onychomycosis (see US 4,755,534 - Sandoz)

[0110] **Docosanol,** also known as behenyl alcohol, is a saturated fatty alcohol used traditionally as an emollient, emulsifier, and thickener in cosmetics, nutritional supplement (as an individual entity and also as a constituent of policosanol), and more recently, in a Food and Drug Administration (FDA) approved pharmaceutical, Abreva, approved as an antiviral agent for reducing the duration of cold sores caused by the herpes simplex virus.

[0111] **Ciclopiroxolamine** (6-cyclohexyl-1-hydroxy-4-methylpyridin-2(1H)-one) also called Batrafen, Loprox, Mycoster, Penlac and Stieprox, is a synthetic antifungal agent for topical dermatologic treatment of superficial mycoses.



[0112] It is most useful against *Tinea versicolor* (see US 3,883,545 - Marck).

Anti-cellulite agents

[0113] Agents enhancing or boosting the activity of anti-cellulite agents, in particular agents which stimulate and/or depolarise C nerve fibres, are preferably selected from the group consisting of capsaicin and derivatives thereof, vanillylnonylamid and derivatives thereof, L-carnitine, coenzym A, isoflavonoides, soy extracts, ananas extract and conjugated linoleic acid.

Fat enhancing agents

[0114] Formulations and products according to the present invention may also comprise one or more fat enhancing and/or adipogenic agents as well as agents enhancing or boosting the activity of fat enhancing agents. A fat enhancing agent is for example hydroxymethoxyphenyl propylmethylmethoxybenzofuran (trade name: Sym3D™).

Hair growth activators or inhibitors

[0115] Formulations and products according to the present invention may also comprise one or more hair growth activators, i.e. agents to stimulate hair growth. Hair growth activators are preferably selected from the group consisting of pyrimidine derivatives such as 2,4-diaminopyrimidine-3-oxide (Aminexil), 2,4-diamino-6-piperidinopyrimidine-3-oxide (Minoxidil) and derivatives thereof, 6-amino-1,2-dihydro-1-hydroxy-2-imino-4-piperidinopyrimidine and its derivatives, xanthine alkaloids such as caffeine, theobromine and theophylline and derivatives thereof, quercetin and derivatives, dihydroquercetin (taxifolin) and derivatives, potassium channel openers, antiandrogenic agents, synthetic or natural 5-reductase inhibitors, nicotinic acid esters such as tocopheryl nicotinate, benzyl nicotinate and C1-C6 alkyl nicotinate, proteins such as for example the tripeptide Lys-Pro-Val, diphencypren, hormons, finasteride, dutasteride, flutamide, bicalutamide, pregnane derivatives, progesterone and its derivatives, cyproterone acetate, spironolactone and other diuretics, calcineurin inhibitors such as FK506 (Tacrolimus, Fujimycin) and its derivatives, Cyclosporin A and derivatives thereof, zinc and zinc salts, polyphenols, procyanidins, proanthocyanidins, phytosterols such as for example beta-sitosterol, biotin, eugenol, (±)-beta-citronellol, panthenol, glycogen for example from mussels, extracts from microorganisms, algae, plants and plant parts of for example the genera dandelion (Leontodon or Taraxacum), Orthosiphon, Vitex, Coffea, Paullinia, Theobroma, Asiasarum, Cucurbita or Styphnolobium, Serenoa repens (saw palmetto), Sophora flavescens, Pygeum africanum, Panicum miliaceum, Cimicifuga racemosa, Glycine max, Eugenia caryophyllata, Cotinus coggygria, Hibiscus rosa-sinensis, Camellia sinensis, Ilex paraguariensis, licorice, grape, apple, barley or hops or/nd hydrolysates from rice or wheat.

[0116] Alternatively, formulations and products according to the present invention may comprise one or more hair growth inhibitors (as described above), i.e. agents to reduce or prevent hair growth. Hair growth inhibitors are preferably selected from the group consisting of activin, activin derivatives or activin agonists, ornithine decarboxylase inhibitors such as alpha-difluoromethylornithine or pentacyclic triterpenes like for example ursolic acid, betulin, betulinic acid, oleanolic acid and derivatives thereof, 5alpha-reductase inhibitors, androgen receptor antagonists, S-adenosylmethionine decarboxylase inhibitors, gamma-glutamyl transpeptidase inhibitors, transglutaminase inhibitors, soybean-derived serine protease inhibitors, extracts from microorganisms, algae, different microalgae or plants and plant parts of for example the families Leguminosae, Solanaceae, Graminae, Asclepiadaceae or Cucurbitaceae, the genera Chondrus, Gloiopeltis, Ceramium, Durvillea, Glycine max, San-guisorba officinalis, Calendula officinalis, Hamamelis virginiana, Arnica montana, Salix alba, Hypericum perforatum or Gymnema sylvestre.

Solutes

[0117] Formulations and products according to the present invention may also comprise one or more compatible solutes. Preferred compatible solutes are such as described in WO 01/76572, particularly dimyo-inositol phosphate (DIP), diglycerin phospate (DGP), di-myoinositol phosphate (DIP), cyclic 2,3 diphosphoglycerate (cDPG), 1,1-di-glycerol phosphate (DGP), beta-mannosyl glycerate (firoin), beta-mannosyl glyceramide (firoin-A) and di-mannosyl-di-inositol phosphate (DMIP) and ectoine and ectoine-derivatives, as described in EP 0 553 884, EP 0 671 161 and WO 94/15923, in particular ((S)-1,4,5,6-tetrahydro-2-methyl-4-pyrimidinecarboxylic acid) and hydroxyectoine ((S,S)-1,4,5,6-tetrahydro-5-hydroxy-2-methyl-4-pyrimidinecarboxylic acid).

[0118] Preferably, the total amount of compatible solutes is in the range of from 0.05 to 10 wt.-%, preferably from 0.1 to 5 wt.-%, based on the total weight of the formulation or product.

Solvents

[0119] The pharmaceutical compositions may contain such as for example aliphatic alcohols or 1,2-alkandiols or of course simply water.

[0120] **1,2-Alkandiols.** Suitable 1,2-alkandiols encompass 1,2-butadiol, 1,2-pentandiol, 1,2-heptanddiol, 1,2-nonandiol, 1,2-decandiol, 1,2-undecandiol, 1,2,dodecandiol and their mixtures. The preferred 1,2-alkandiol is 1,2-pentandiol.

[0121] **Aliphatic alcohols.** Suitable aliphatic alcohols are selected from the group consisting of ethanol, n-propanol, isopropylalcohol, the isomeric butanols and their mixtures. The preferred species is ethanol, in particular with a purity of at least 95 %.

INDUSTRIAL APPLICATION

Additional objects of the present invention refer to a method for

[0122]

(i) improving the stability of an emulsion and/or

(ii) decreasing the average particle distribution of droplets in an emulsion, and/or

(iii) improving the sensory profile of an emulsion,

by adding a working amount of the mixture 1.2-hexanediol and 1,8-hexanediol,

- preferably in a ratio by weight of from about 25:75 to about 75:25, more preferably in an amount by weight of from about 40:60 to about 60:40 and particularly in an amount by weight of about 50:50,
- wherein said working amount is about 0.1 to about 1.0 wt.-% and preferably about 0.2 to about 0.5 wt.-% - calculated on the final emulsion.

[0123] Finally, another object of the present invention is related to the use of binary mixtures of 1,2-hexanediol and, 2-octanediol, preferably in a ratio by weight as disclosed above for simultaneously improving stability, decreasing average particle size distribution and improving sensory profile of an emulsion, wherein said binary mixture is preferably to said emulsion in an amount of from about 0.1 to about 1.0 wt.-% and preferably about 0.2 to about 0.5 wt.-% - calculated on the final emulsion.

EXAMPLES

EXAMPLE 1, COMPARATIVE EXAMPLE C1 TO C2

Antimicrobial activity

[0124] Antimicrobial activity of a mixture of 1,2-hexanediol and 1,2-octanediol (1:1) was investigated compared to the individual 1,2-diols with regard to 5 prominent microorganisms:

EC *Echerichia coli*

PA *Pseudomonas aeruginosa*
SA *Staphylococcus aureus*
CA *Candida albicans*
AN *Aspergillus niger*

[0125] The results are presented in **Table 1.** Shown are the minimum inhibitory concentrations (MICs) in percent.

Table 1

Antimicrobial activity							
Ex.	**1.2-hexanediol**	**1,2-octanediol**	**EC**	**PA**	**SA**	**CA**	**AN**
1	50.0	50.0	0.31	0.60	0.31	0.16	0.08
C1	100.0	-	1.25	0.63	2.50	1.25	0.63
C2	-	100.00	0.63	0.63	1.25	0.31	0.16

[0126] The examples proof that the mixture according to the present invention shows a synergism with regard to the minimal concentration for inhibiting growth of 4 out of 5 microorganisms; for Pseudomonas only a small reduction was achieved.

EXAMPLE 2, COMPARATIVE EXAMPLES C3 TO C5

Particle size distribution

[0127] O/W emulsions were prepared by heating a lipid phase A and an aqueous phase B separately to approximately 80 °C. Then the aqueous phase B was added to the lipid phase A and homogenized by using an Ultra-Turrax Stirrer for 2 minutes at 6.000 rpm. The emulsion thus obtained was allowed to cool down for 10 minutes using a vane stirrer at 150 rpm. Finally, the pH value was adjusted to about 6.0 by adding aqueous sodium hydroxide solution. The particle size of the resulting emulsions was determined using a Mastersizer Micro MAF 500 (Malvern) using the principle of laser diffraction. The compositions and average particle sizes are shown in **Table 2.** Comparative Example C2 is a placebo, Comparative Examples C3 and C4 were prepared with the individual 1,2-diols. Example 2 is according to the invention.

Table 2
Composition of o/w emulsions (all amounts in wt.-%)

Ph.	Raw Material	INCI	C2	2	C3	C4
A.	Emulsiphos® PN677660	Potassium Cetyl Phosphate, Hydrogenated Palm Glycerides	2,0	2,0	2,0	2,0
	Lanette O, ex BASF	Cetearyl Alcohol	0,7	0,7	0,7	0,7
	Neutral Oil	Caprylic/Capric Triglyceride	8,0	8,0	8,0	8,0
	PCL Liquid 100 PN660089	Cetearyl Ethylhexanoate	4,0	4,0	4,0	4,0
	Abil 350, ex Evonik	Dimethicone	0,1	0,1	0,1	0,1
B.	1,2 Hexanediol	1,2 Hexanediol	-	0,25	0,5	-
	1,2 Octanediol	Caprylyl Glycol	-	0,25	-	0,5
	Water, demin.	Water (Aqua)	83,7	83,2	83,2	83,2
	Glycerin 99%	Glycerin	1,5	1,5	1,5	1,5
Particle size - all values in µm (mean values, double determination)						
	D (v, 0.1) = 10 %		1,7	1,5	1,7	2,0
	D (v, 0.5) = 50 %		8,9	3,8	8,6	16,3
	D (v, 0.9) = 90 %		40,0	12,1	43,1	50,1

[0128] With regard to the D(v, 0.5) and the D(v, 0.9) values the respective Synergy Index (SI) was calculated according to Kull's equation. Synergy in a composition exists when the synergy index is less than 1, means the lower the SI the greater the synergy.

- SI for D(v, 0.5) value = 0,337
- SI for D(v, 0.9) = 0,261

[0129] The particle size distribution for Example 2 and the Comparative Examples C2 to C4 is also depicted in **Figure 1**. The index "v" refers to a volume based particle size distribution; all values were taken from the sum distribution Q_r. For example, the value 1.7 for C2 means that 10 % of the particles show a particle size less than 1.7 µm and so on. As one can see, adding the mixture of 1,2-hexanediol and 1,2-octanediol shifts the average particle size to significant smaller values and leads to more finely divided emulsions in comparison to 1,2-hexanediol and 1,2-octanediol alone. The highly synergistic effect could be clearly demonstrated and calculated.

[0130] **Figure 2** provides microscopic images of the 4 compositions. The samples were observed via microscope. (Olympus IX 70, 600 fold). Figure 2B (= Example 2), emulsion with a mixture of 1,2-hexanediol and 1,2-octanediol, showed significant smaller particles in comparison to 1,2-hexanediol and 1,2-octanediol alone (Figures 2C and 2D) and therefore supports the results of particle size measurements.

Claims

1. An active mixture comprising

   (a) 1,2-hexanediol and

(b) 1,2-octanediol.

2. The composition of Claim 1, wherein said components (a) and (b) are present in a ratio by weight of from about 25:75 to about 75:25.

3. The composition of Claim 1, wherein said components (a) and (b) are present in a ratio by weight of from about 40:60 to about 60:40.

4. A cosmetic composition comprising the mixture of Claim 1.

5. A pharmaceutical compositions comprising the mixture of Claim 1.

6. The composition of Claim 4 being a skin care, hair care, sun care or personal care composition.

7. The composition of Claim 4 or Claim 5 being an emulsion.

8. The emulsion of Claim 7, being an oil-in-water (o/w) emulsion, a water-in-oil (w/o) emulsion or a multiple (w/o/w or o/w/o) emulsion.

9. The composition of Claim 4 or Claim 5, being an emulsion with an average particle size, where 10 Vol.-% show a diameter of less than 2 μm, 50 Vol.-% show a diameter of less than 5 μm and 90 Vol.-% show a diameter of less than 15 μm.

10. A method for improving the stability of an emulsion by adding a working amount of the mixture of Claim 1.

11. A method for decreasing the average particle distribution of droplets in an emulsion by adding a working amount of the mixture of Claim 1.

12. A method for improving the sensory profile of an emulsion by adding a working amount of the mixture of Claim 1.

13. The method of Claim 10, 11 or 12, wherein said working amount is about 0.1 to about 1.0 wt.-% - calculated on the final emulsion.

14. Use of the mixture of Claim 1 for simultaneously improving stability, decreasing average particle size distribution and improving sensory profile of an emulsion.

15. The use of Claim 14, wherein said mixture is added to said emulsion in an amount of from about 0.1 to about 1 wt.-% - calculated on the final emulsion.

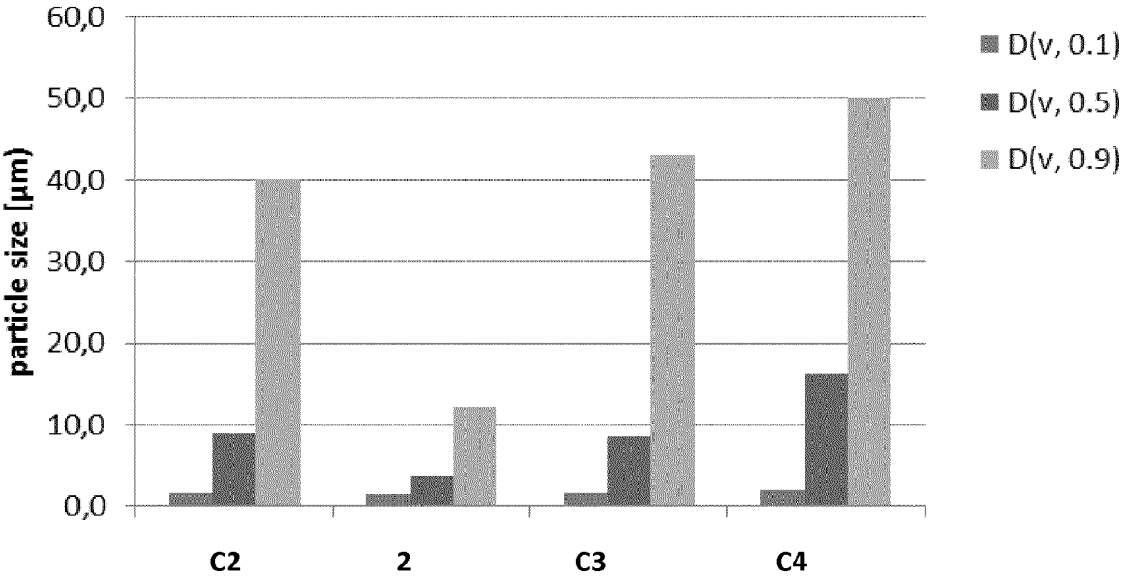
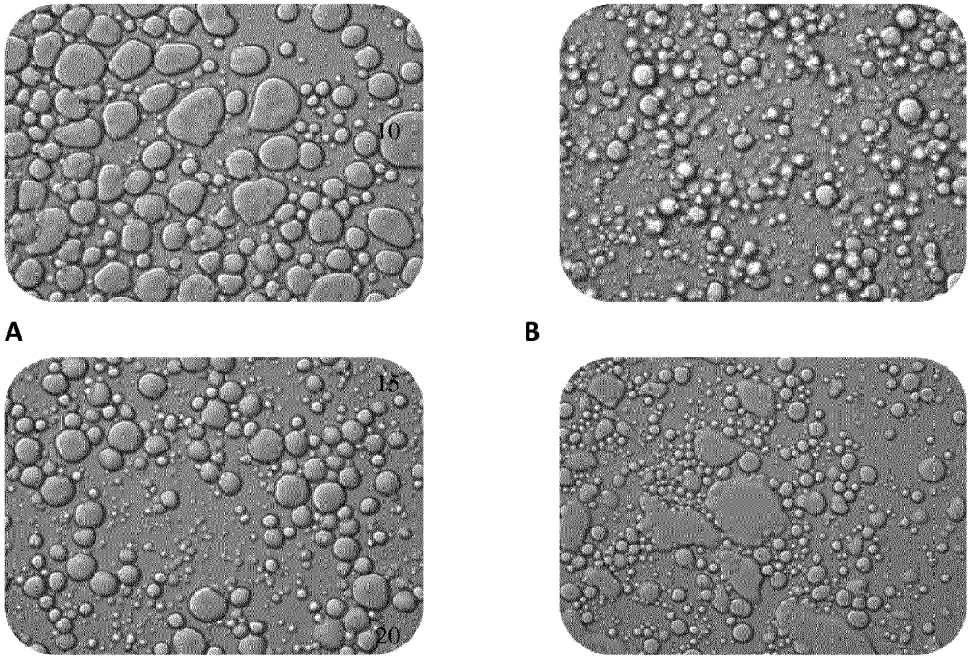


Figure 1



A B

C D

Figure 2

Europäisches Patentamt
European Patent Office
Office européen des brevets

EUROPEAN SEARCH REPORT

Application Number
EP 15 15 1564

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (IPC)
X,D	WO 03/069994 A1 (DRAGOCO GERBERDING CO AG [DE]; SCHMAUS GERHARD [DE]; LANGE SABINE [DE]) 28 August 2003 (2003-08-28) * claims; examples; table 8 * * page 26, paragraph 3 * * page 12, paragraph 5 - page 13, paragraph 1 *	1-15	INV. A61K8/34 A61K8/06 A61Q19/00
X	DE 10 2008 043586 A1 (SYMRISE GMBH & CO KG [DE]) 14 May 2009 (2009-05-14) * example 15 *	1-15	
X	RAVI PILLAI ET AL: "1, 2-Alkanediols for Cosmetic Preservation", COSMETICS & TOILETRIES, WHEATON, IL, US, vol. 123, October 2008 (2008-10), pages 53-64, XP009131860, ISSN: 0361-4387 * page 63, last paragraph - page 64, column 1; table 4 *	1-15	
X	WO 2006/069953 A1 (SYMRISE GMBH & CO KG [DE]; SCHMAUS GERHARD [DE]; PILLAI RAVIKUMAR [US]) 6 July 2006 (2006-07-06) * claim 3; examples *	1-15	TECHNICAL FIELDS SEARCHED (IPC) A61Q A61K
X	ZhangWei: "Study on the Non-aqueous Polar Media Microemulsion Systems Containing Vitamin C", East China University of Science and Technology , 2012, XP002749044, Retrieved from the Internet: URL:http://www.dissertationtopic.net/doc/29861 [retrieved on 2015-10-27] * the whole document *	1-15	

-/--

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
Munich	28 October 2015	Blott, Catherine

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
Y : particularly relevant if combined with another document of the same category
A : technological background
O : non-written disclosure
P : intermediate document

T : theory or principle underlying the invention
E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document

EPO FORM 1503 03.82 (P04C01)

Europäisches Patentamt
European Patent Office
Office européen des brevets

EUROPEAN SEARCH REPORT

Application Number
EP 15 15 1564

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (IPC)
X	WO 2014/135650 A1 (SYMRISE AG [DE]; PESARO MANUEL [DE]; HÖLSCHER BERND [DE]; SCHMAUS GERH) 12 September 2014 (2014-09-12) * paragraph [0292] *	1-15	
X	SCHMAUS G ET AL: "SYNERGISTIC COMBINATION WITH MULTIPLE FUNCTIONALITY COMPRISING PHENOXYETHANOL AND 1,2-ALKANEDIOLS", 27TH IFSCC CONGRESS 2012, SOUTH AFRICA - BEAUTY IN DIVERSITY - A GLOBAL VILLAGE, 16-18 OCTOBER 2012, SANDTON CONVENTION CENTER, GAUTENG, SOUTH AFRICA, BOOK OF ABSTRACTS,, 16 October 2012 (2012-10-16), page 5pp, XP009185355, ISBN: 978-0-620-53667-7 * the whole document *	1-15	
E	WO 2015/063471 A1 (DEB IP LTD [GB]) 7 May 2015 (2015-05-07) * page 21, line 2 - line 7; claims 1,7,9,10; examples *	1-8, 10-15	

TECHNICAL FIELDS SEARCHED (IPC)

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
Munich	28 October 2015	Blott, Catherine

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
Y : particularly relevant if combined with another document of the same category
A : technological background
O : non-written disclosure
P : intermediate document

T : theory or principle underlying the invention
E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document

EPO FORM 1503 03.82 (P04C01)

ANNEX TO THE EUROPEAN SEARCH REPORT ON EUROPEAN PATENT APPLICATION NO. EP 15 15 1564

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on
The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

28-10-2015

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 03069994 A1	28-08-2003	AT 337703 T	15-09-2006
		AU 2002350557 A1	09-09-2003
		CN 1620248 A	25-05-2005
		DE 10206759 A1	28-08-2003
		EP 1478231 A1	24-11-2004
		EP 1731036 A1	13-12-2006
		ES 2269781 T3	01-04-2007
		JP 4852231 B2	11-01-2012
		JP 2005526036 A	02-09-2005
		US 2005222276 A1	06-10-2005
		WO 03069994 A1	28-08-2003
DE 102008043586 A1	14-05-2009	DE 102008043586 A1	14-05-2009
		US 2009123392 A1	14-05-2009
WO 2006069953 A1	06-07-2006	EP 1835843 A1	26-09-2007
		JP 2008525518 A	17-07-2008
		US 2010216892 A1	26-08-2010
		WO 2006069953 A1	06-07-2006
WO 2014135650 A1	12-09-2014	EP 2774481 A1	10-09-2014
		JP 2014172908 A	22-09-2014
		WO 2014135650 A1	12-09-2014
WO 2015063471 A1	07-05-2015	DE 102013222164 A1	30-04-2015
		WO 2015063471 A1	07-05-2015

EPO FORM P0459

For more details about this annex : see Official Journal of the European Patent Office, No. 12/82

REFERENCES CITED IN THE DESCRIPTION

This list of references cited by the applicant is for the reader's convenience only. It does not form part of the European patent document. Even though great care has been taken in compiling the references, errors or omissions cannot be excluded and the EPO disclaims all liability in this regard.

Patent documents cited in the description

- WO 2003069994 A1 **[0006]**
- DE 10055940 A1 **[0041]**
- WO 2002038537 A1 **[0041]**
- DE 10055940 **[0042]**
- WO 0238537 A **[0042]**
- WO 2005123101 A1 **[0043] [0046] [0055]**
- WO 2010122178 A **[0048]**
- WO 2010097480 A **[0048]**
- EP 0584178 A **[0050]**
- WO 2005032501 A **[0050]**
- WO 2005102252 A **[0050]**
- WO 2006010661 A **[0050]**
- WO 0206992 A1 **[0055]**
- WO 2004047833 A1 **[0058] [0059]**
- WO 2009087242 A1 **[0062]**
- EP 0852949 A **[0063]**
- FR 2570377 A1 **[0064]**
- EP 0199636 A1 **[0064]**
- EP 0325540 A1 **[0064]**
- EP 0402072 A1 **[0064]**
- EP 1529522 A1 **[0065]**
- WO 2007077541 A **[0066]**
- WO 2010097479 A **[0066]**
- US 4150052 A **[0070]**
- WO 2005049553 A1 **[0070]**
- WO 2004026840 A **[0070]**
- EP 2033688 A2 **[0070]**
- DE 10143434 **[0071]**
- DE 4447361 **[0071]**
- DE 10330697 **[0071]**
- US 4110430 A **[0071]**
- EP 1157687 A **[0071]**
- US 2653899 A, Eli Lily **[0092]**
- US 3053865 A, Merck **[0095]**
- US 3385886 A, Boots **[0097]**
- US 3641127 A **[0099]**
- US 3558690 A **[0101]**
- US 2944061 A **[0103]**
- US 4199574 A **[0105]**
- US 4689338 A, Riker **[0107]**
- US 4755534 A, Sandoz **[0109]**
- US 3883545 A, Marck **[0112]**
- WO 0176572 A **[0117]**
- EP 0553884 A **[0117]**
- EP 0671161 A **[0117]**
- WO 9415923 A **[0117]**

Non-patent literature cited in the description

- Surfactants in Consumer Products. Springer Verlag, 1987, 54-124 **[0016]**
- Katalysatoren, Tenside und Mineralölladditive. Thieme Verlag, 1978, 123-217 **[0016]**
- **TODD et al.** *Cosm. Toil.,* 1976, vol. 91, 27 **[0031]**

Electronic Patent Application Fee Transmittal

Application Number:	16262902
Filing Date:	30-Jan-2019
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Filer:	Kathryn Anne Warner/Valerie Foster
Attorney Docket Number:	INLX.0198U2/484799

Filed as Large Entity

Filing Fees for Utility under 35 USC 111(a)

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Pages:				
Claims:				
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
Extension-of-Time:				

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Extension - 1 month with $0 paid	1251	1	220	220
Miscellaneous:				
RCE- 2ND AND SUBSEQUENT REQUEST	1820	1	2000	2000
		Total in USD ($)		**2220**

Electronic Acknowledgement Receipt

EFS ID:	45243659
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	171052
Filer:	Kathryn Anne Warner/Valerie Foster
Filer Authorized By:	Kathryn Anne Warner
Attorney Docket Number:	INLX.0198U2/484799
Receipt Date:	16-MAR-2022
Filing Date:	30-JAN-2019
Time Stamp:	15:51:33
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	DA
Payment was successfully received in RAM	$2220
RAM confirmation Number	E20223FF52204151
Deposit Account	
Authorized User	

The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows:

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1		INLX_0198U2_484799_Amendment_and_RCE.pdf	227647 41ca37010b71bdac5b112e590d84bbef02773c82	yes	7

Multipart Description/PDF files in .zip description

Document Description	Start	End
Response After Final Action	1	1
Claims	2	4
Applicant Arguments/Remarks Made in an Amendment	5	7

Warnings:

Information:

2	Extension of Time	INLX_0198U2_484799_ExtensionOfTime.pdf	166422 cca2fc5d88063deb0a678ffa091638532ca08beb	no	2

Warnings:

Information:

3	Request for Continued Examination (RCE)	INLX_0198U2_484799_Request_for_Continued_Examination.pdf	1670491 b23ace72bb30699c991649cd2200450d2a302a30	no	3

Warnings:

Information:

4	Information Disclosure Statement (IDS) Form (SB08)	INLX_0198U2_484799_Information_Disclosure_Statement.PDF	1284969 acbaa3a80b5f562a1cd61447818d80490265091c	no	4

Warnings:

Information:

5	Non Patent Literature	INX_0198U2_484799_SupplementaryEuropeanSearchReport.pdf	431628 016d322e18c6f7c62967e3d8bf4efae9f327fa18	no	12

Warnings:					
Information:					
6	Foreign Reference	EP3045161A1_Symrise_AG.PDF	3033182 0a42d9669ae9a4b5038feb2ba541a47ff96f5aea	no	32
Warnings:					
Information:					
7	Fee Worksheet (SB06)	fee-info.pdf	41126 f5d107f4724b3cdb9ae98bf117a6a29b45802fb4	no	2
Warnings:					
Information:					
			Total Files Size (in bytes):	6855465	

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor: **Rocco V. Burgo**	Attorney Docket No: **INLX.0198U2/484799**
Application No.: **16/262,902**	Confirmation No.: **1028**
Filing Date: **January 20, 2019**	Art Unit: **1612**
Title: ***Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same***	Examiner: **HOLLOMAN, Nannette**

VIA EFS

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

AMENDMENT AND REQUEST FOR CONTINUED EXAMINATION

UNDER 37 C.F.R. § 1.114

This amendment and response is in answer to the Final Office Action mailed December 1, 2021. A one month extension of time fee is included with this request. Applicants hereby authorize the Commissioner to charge any other necessary fees, including fees for extensions of time or claims, to Deposit Account No. 50-3111. Applicants respectfully request reconsideration of this application in view of the following claim amendments and remarks:

A marked-up **Listing of the Claims** begin on page 2.

Remarks begin on page 5.

LISTING OF THE CLAIMS

This listing of claims will replace all prior versions, and listings, of claims in the application.

What is Claimed is:

1-41. (Canceled)

42. (Currently Amended) A preservative composition, comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of 7 to 20 carbon atoms, wherein the carbon atoms of the [[first]] bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived, and wherein the bio-1,2-alkanediol is prepared from a bio-1-alkene having 7 to 20 carbon atoms.

43. (Previously Presented) A personal care composition comprising the preservative composition of claim 42.

44. (Previously Presented) The personal care composition of claim 43, wherein the personal care composition is a hair care composition, an oral care composition, a skin care composition or a cosmetic composition.

45. (Previously Presented) The personal care composition of claim 43, wherein the personal care composition is a household product.

46. (Previously Presented) The personal care composition of claim 45, wherein the household product is a fabric care product or a cleaning product.

47. (Previously Presented) The personal care composition of claim 43, wherein the bio-1,2-alkanediol is selected from the group consisting of: bio-1,2-heptanediol, bio-1,2-octanediol, bio-1,2-decanediol, bio-1,2-dodecanediol, and combinations thereof.

48. (Previously Presented) The personal care composition of claim 43, wherein the personal care composition is a pharmaceutical composition, a vitamin composition, or a health care composition.

49. (Previously Presented) The personal care composition of claim 43, wherein the preservative composition is present in a concentration of about 0.1% to about 10%.

50. (Previously Presented) The personal care composition of claim 43, further comprising a bio-compound selected from the group consisting of: a bio-organic acid, bio-hydroxamic acid, a bio-alkylhydroxamic acid, a bio-terpenoid compound having 5 to 20 carbon

atoms, bio-1,3-propanediol, bio-1,2-butanediol, bio-1,2-pentanediol, and combinations thereof.

51. (Previously Presented) The personal care composition of claim 50, wherein the preservative composition is present in a concentration of about 0.1% to about 10%.
52. (Previously Presented) The preservative composition of claim 42, wherein the composition further comprises at least one other bio-compound different from the bio-1,2-alkanediol.
53. (Previously Presented) The preservative composition of claim 52, further comprising an antimicrobial compound different from the-bio-1,2-alkanediol.
54. (Previously Presented) The preservative composition of claim 42, comprising about 75% to about 99% of the bio-1,2-alkanediol.
55. (Previously Presented) The preservative composition of claim 42, wherein the bio-1,2-alkanediol comprises about 60% to about 99% bio-1,2-octanediol.

56-71. (Canceled).

72. (Currently Amended) A preservative composition comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of 7 to 14 carbon atoms, wherein the carbon atoms of the bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived, and wherein the bio-1,2-alkanediol is prepared from a bio-alkene having 7 to 14 carbon atoms.
73. (Previously Presented) The preservative composition of claim 72, wherein the bio-1,2-alkanediol is selected from the group consisting of: bio-1,2-heptanediol, bio-1,2-octanediol, bio-1,2-decanediol, bio-1,2-dodecanediol, and combinations thereof.
74. (Previously Presented) The preservative composition of claim 72, wherein the bio-1,2-alkanediol comprises about 60% to about 99% of bio-1,2-octanediol.
75. (Previously Presented) The preservative composition of claim 72, wherein the bio-1,2-alkanediol comprises about 75% to about 99% bio-1,2-octanediol.
76. (Previously Presented) The preservative composition of claim 72, further comprising a bio-compound selected from the group consisting of: a bio-organic acid, bio-hydroxamic acid, a bio-alkylhydroxamic acid, a bio-terpenoid compound having 5 to 20 carbon atoms, bio-1,3-propanediol, bio-1,2-butanediol, bio-1,2-pentanediol, bio-1,2-hexanediol, and combinations thereof.

77. (Previously Presented) A personal care composition comprising the preservative composition of claim 72 in a concentration of about 0.1% to about 10%.

78. (Previously Presented) The personal care composition of claim 77, wherein the preservative composition is present in a concentration of about 0.3% to about 2%.

79. (Previously Presented) The personal care composition of claim 78, wherein the preservative composition comprises about 60% to about 99% of bio-1,2-octanediol.

80. (Previously Presented) The personal care composition of claim 78, wherein the preservative composition comprises about 75% to about 99% of bio-1,2-octanediol.

81-87. (Canceled).

88. (New) A preservative composition, comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of 7 to 20 carbon atoms, wherein the carbon atoms of the bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived, and wherein the preservative composition comprises less than 20% of a corresponding bio-2,3-alkanediol having the same carbon chain length as the bio-1,2-alkanediol.

89. (New) The preservative composition of claim 88, wherein the bio-1,2-alkanediol has a carbon chain length of 7 to 14 carbon atoms.

90. (New) The preservative composition of claim 89, comprising less than about 8% of the corresponding bio-2,3-alkanediol having the same carbon chain length as the bio-1,2-alkanediol.

91. (New) The preservative composition of claim 90, comprising less than about 5% of the corresponding bio-2,3-alkanediol having the same carbon chain length as the bio-1,2-alkanediol.

REMARKS

Application and Claims Status

Upon entry of the foregoing claim amendments, claims 42-55, 72-80, and 88-91 are pending. Claims 1-41, 56-71, and 81-87 were previously cancelled.

Claims 42 and 72 are amended, and claims 88-91 are added. Written support for the amendments to claims 42 and 72, and new claims 88-91, can be found in the as-filed specification at, *inter alia*, ¶¶ [0029]-[0030], [0037], [0061], [0065]-[0067], [0070], [0091]-[0092], [0126]-[0130], FIG. 1, and the Abstract.

Claims 42-55 and 72-80 are rejected. In view of the following remarks, Applicants respectfully submit that the pending rejections have been traversed and respectfully request favorable reconsideration and allowance of the pending claims.

Examiner Interview

Applicants thank the Examiner for taking time to speak with their undersigned counsel of record and Mike Fevola, Ph.D. on Wednesday, January 19, 2022. During the interview, the prior art rejection was discussed, as well as possible claim amendments that may place this application into allowable form. Aspects of the discussion are incorporated into the above claim amendments and following remarks.

Patentability of the Claims

The Rejections For Obviousness Under 35 U.S.C. § 103 Should be Withdrawn

Claims 42-44, 47-55, and 72-80 were rejected under 35 U.S.C. § 103 as allegedly obvious over Schmaus (US Patent Pub. No. 2005/0222276) in view of Wang (*Green Chem.*, 14:758-65 (2012). Final Office Action at 2-4.

Claims 42-55 and 72-80 were rejected under 35 U.S.C. § 103 as allegedly obvious over Lutz (US Patent Pub. No. 2012/0282207) in view of Wang. Final Office Action at 4-5.

Based on the foregoing claim amendments, Applicants respectfully submit that claims 42-55 and 72-80 are in condition for allowance. Specifically, during the interview of January 19, 2022, the Examiner expressed that claim amendments reflective of the process by which the bio-1,2-alkanediol compounds are prepared should place the claims in condition for allowance. Independent claims 42 and 72 have been amended to require "wherein the bio-1,2-alkanediol is

prepared from a bio-1-alkene having 7 to 20 carbon atoms," and "wherein the bio-1,2-alkanediol is prepared from a bio-1-alkene having 7 to 14 carbon atoms," respectively. As such, claims 42 and 72, and claims 43-55 and 73-80 that depend therefrom, all recite the process by which the claimed bio-1,2-alkanediol compositions are prepared.

As noted in Applicants' Response dated November 11, 2021, neither Schmaus nor Lutz teach or suggest how to prepare any bio-1,2-alkanediols, let alone bio-1,2-alkanediols having a chain length of 7 to 20 carbon atoms, let alone through a bio-1-alkene intermediate. Wang does not remedy this deficiency, as Wang's disclosure is limited to methods of using cellulosic feedstocks to prepare bio-C_2-C_6 alkanediols,. *See* Wang at 761, Table 1. As such, a person of ordinary skill in the art following Schmaus or Lutz in view of Wang would have no way to make any 1,2-alkanediols through a bio-1-alkene intermediate. For these reasons, Applicants respectfully request reconsideration and withdrawal of the rejections over Schmaus in view of Wang and Lutz in view of Wang.

New Claims 88-91 Are Similarly Non-Obvious Over the Prior Art of Record

Claims 88-91 require, *inter alia*, "[a] preservative composition, comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of 7 to 20 carbon atoms, . . . wherein the preservative composition comprises less than 20% of a corresponding bio-2,3-alkanediol having the same carbon chain length as the bio-1,2-alkanediol." As discussed in the as-filed specification, the disclosed process for preparing bio-1,2-alkanediols utilizes a bio-1-alkene intermediate having a regioselectivity of at least 80%. *See, e.g.*, ¶¶ [0065]-[0067]. This regioselectivity importantly minimizes the formation of the corresponding bio-2,3-alkanediol impurity. *See, e.g.*, ¶¶ [0126]-[0128] (Examples 5-7, describing a bio-1-alkene having a regioselectivity of 96%, which then provides a bio-1,2-alkanediol purity of 98.7%). That is, the conversion of the bio-1-alkene to the corresponding claimed bio-1,2-alkanediol occurs with near unity yield, and therefore, the bio-1-alkene regioselectivity is largely responsible for providing the purity of the resulting claimed bio-1,2-alkanediol composition. Thus, Applicants respectfully submit that new claims 88-91, which require "less than 20% of a corresponding bio-2,3-alkanediol having the same carbon chain length as the bio-1,2-alkanediol," are reflective of the process disclosed in the instant application.

For these reasons, Applicants respectfully submit that new claims 88-91 are in condition for allowance.

CONCLUSION

In view of the foregoing claim amendments and remarks, this Request for Continued Examination places this application in condition for allowance, and therefore, its entry is proper under 37 C.F.R. § 1.114. Applicants respectfully submit that the pending claims are in condition for allowance and Notice of Allowance is earnestly solicited. The Examiner is invited to contact the undersigned agent with any questions, comments or suggestions relating to the referenced patent application.

This response is being submitted on March 16, 2022. In the event the Commissioner determines that any fees are required for any reason, the Commissioner is hereby authorized to deduct said fee from Deposit Account No.: 50-3111/Matter No.: 484799.000.

Respectfully submitted,

Date: March 16, 2022

/kathryn a warner/

Kathryn A. Warner
Reg. No. 59,858
COZEN O'CONNOR
1200 Nineteenth Street N.W.
Washington, D.C. 20036
Tel: (720) 479-3878
Fax: (202) 861-1905

PTO/AIA/22 (10-20)
Approved for use through 11/30/2020. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

PETITION FOR EXTENSION OF TIME UNDER 37 CFR 1.136(a)

Docket Number (Optional): INLX.0198U2/484799

Application Number: 16/262,902

Filed: 2019-01-30

For: Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same

Art Unit: 1612

Examiner: HOLLOMAN, Nannette

This is a request under the provisions of 37 CFR 1.136(a) to extend the period for filing a reply in the above-identified application.

The requested extension and fee are as follows (check time period desired and enter the appropriate fee below):

		Fee	Small Entity Fee	Micro Entity Fee	
☑	One month (37 CFR 1.17(a)(1))	$220	$110	$55	$ 220.00
☐	Two months (37 CFR 1.17(a)(2))	$640	$320	$160	$
☐	Three months (37 CFR 1.17(a)(3))	$1,480	$740	$370	$
☐	Four months (37 CFR 1.17(a)(4))	$2,320	$1,160	$580	$
☐	Five months (37 CFR 1.17(a)(5))	$3,160	$1,580	$790	$

☐ Applicant asserts small entity status. See 37 CFR 1.27.

☐ Applicant certifies micro entity status. See 37 CFR 1.29.
Form PTO/SB/15A or B or equivalent must either be enclosed or have been submitted previously.

☐ A check in the amount of the fee is enclosed.

☐ Payment by credit card. Form PTO-2038 is attached.

☐ The Director has already been authorized to charge fees in this application to a Deposit Account.

☑ The Director is hereby authorized to charge any fees which may be required, or credit any overpayment, to Deposit Account Number 50-3111.

☑ Payment made via EFS-Web.

WARNING: Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.

I am the

☐ applicant.

☑ attorney or agent of record. Registration number 59,858.

☐ attorney or agent acting under 37 CFR 1.34. Registration number _____.

/kathryn a warner/ — Signature

March 16, 2022 — Date

Kathryn A. Warner — Typed or printed name

(202) 912-4800 — Telephone Number

NOTE: This form must be signed in accordance with 37 CFR 1.33. See 37 CFR 1.4 for signature requirements and certifications. Submit multiple forms if more than one signature is required, see below*.

☑ * Total of 1 forms are submitted.

This collection of information is required by 37 CFR 1.136(a). The information is required to obtain or retain a benefit by the public, which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.11 and 1.14. This collection is estimated to take 6 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Mail Stop PCT, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.

Privacy Act Statement

The **Privacy Act of 1974 (P.L. 93-579)** requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether disclosure of these records is required by the Freedom of Information Act.
2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.
3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.
4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).
5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.
6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).
7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (*i.e.*, GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.
8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspection or an issued patent.
9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	INLX.0198U2/484799	1028

171052 7590 03/30/2022

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

EXAMINER
HOLLOMAN, NANNETTE

ART UNIT	PAPER NUMBER
1612	

NOTIFICATION DATE	DELIVERY MODE
03/30/2022	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

nyipdocketing@cozen.com
vdotson@cozen.com

Office Action Summary	Application No. 16/262,902	Applicant(s) Burgo et al.	
	Examiner NANNETTE HOLLOMAN	Art Unit 1612	AIA (FITF) Status Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTHS FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

1)☑ Responsive to communication(s) filed on 03/16/2022.

☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____.

2a)☐ This action is **FINAL.** 2b) ☑ This action is non-final.

3)☐ An election was made by the applicant in response to a restriction requirement set forth during the interview on _____; the restriction requirement and election have been incorporated into this action.

4)☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims*

5) ☑ Claim(s) 42-55,72-80 and 88-91 is/are pending in the application.

5a) Of the above claim(s) _____ is/are withdrawn from consideration.

6) ☑ Claim(s) 88-91 is/are allowed.

7) ☑ Claim(s) 42-55 and 72-80 is/are rejected.

8) ☐ Claim(s) _____ is/are objected to.

9) ☐ Claim(s) _____ are subject to restriction and/or election requirement

* If any claims have been determined allowable, you may be eligible to benefit from the **Patent Prosecution Highway** program at a participating intellectual property office for the corresponding application. For more information, please see http://www.uspto.gov/patents/init_events/pph/index.jsp or send an inquiry to **PPHfeedback@uspto.gov.**

Application Papers

10)☐ The specification is objected to by the Examiner.

11)☐ The drawing(s) filed on _____ is/are: a)☐ accepted or b)☐ objected to by the Examiner.

Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).

Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).

Priority under 35 U.S.C. § 119

12)☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

Certified copies:

a)☐ All b)☐ Some** c)☐ None of the:

1.☐ Certified copies of the priority documents have been received.

2.☐ Certified copies of the priority documents have been received in Application No. _____.

3.☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

** See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

1) ☑ Notice of References Cited (PTO-892)

2) ☑ Information Disclosure Statement(s) (PTO/SB/08a and/or PTO/SB/08b) Paper No(s)/Mail Date _____.

3) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date _____.

4) ☐ Other: _____.

DETAILED ACTION

Notice of Pre-AIA or AIA Status

The present application, filed on or after March 16, 2013, is being examined under the first inventor to file provisions of the AIA.

This Office Action is in response to the Request for Continued Examination filed on 03/16/2022. Applicants' arguments, filed 03/16/2022, have been fully considered. Rejections and/or objections not reiterated from previous office actions are hereby withdrawn. The following rejections and/or objections are either reiterated or newly applied. They constitute the complete set presently being applied to the instant application.

The text of those sections of Title 35, U.S. Code not included in this action can be found in a prior Office Action.

Allowable Subject Matter

Claims 88-91 appear to be free from art.

Claim Rejections - 35 USC § 103

In the event the determination of the status of the application as subject to AIA 35 U.S.C. 102 and 103 (or as subject to pre-AIA 35 U.S.C. 102 and 103) is incorrect, any correction of the statutory basis for the rejection will not be considered a new ground of rejection if the prior art relied upon, and the rationale supporting the rejection, would be the same under either status.

The following is a quotation of 35 U.S.C. 103 which forms the basis for all obviousness rejections set forth in this Office action:

> A patent for a claimed invention may not be obtained, notwithstanding that the claimed invention is not identically disclosed as set forth in section 102, if the differences between the claimed invention

> and the prior art are such that the claimed invention as a whole would have been obvious before the effective filing date of the claimed invention to a person having ordinary skill in the art to which the claimed invention pertains. Patentability shall not be negated by the manner in which the invention was made.

The factual inquiries for establishing a background for determining obviousness under 35 U.S.C. 103 are summarized as follows:

1. Determining the scope and contents of the prior art.

2. Ascertaining the differences between the prior art and the claims at issue.

3. Resolving the level of ordinary skill in the pertinent art.

4. Considering objective evidence present in the application indicating obviousness or nonobviousness.

This application currently names joint inventors. In considering patentability of the claims the examiner presumes that the subject matter of the various claims was commonly owned as of the effective filing date of the claimed invention(s) absent any evidence to the contrary. Applicant is advised of the obligation under 37 CFR 1.56 to point out the inventor and effective filing dates of each claim that was not commonly owned as of the effective filing date of the later invention in order for the examiner to consider the applicability of 35 U.S.C. 102(b)(2)(C) for any potential 35 U.S.C. 102(a)(2) prior art against the later invention.

Claims 42-44, 47-55 and 72-80 are rejected under 35 U.S.C. 103 as being unpatentable over Schmaus et al. (US Patent Pub. 2005/0222276) in view of Swern et al. (Hydroxylation and Epoxidation of Some 1-Olefins with Per-acids, Journal of the American Chemical Society, Vo. 68, pp 1504-1507, 1946) and Wang et al. (Green Chem., 2012, 14, pp. 758-765, previously disclosed).

Schmaus et al. disclose the use of mixtures of two, three or more 1,2-alkanediols with chain links of 5 to 10 as antimicrobial active compounds (ABSTRACT). Schmaus et al. disclose the use of a mixture

of 1,2-hexanediol and 1,2-octandediol; herein the proportion of each individual diol should be in the range of 1 to 99% (m/m) ([0032] and [0033]). Schmaus et al. disclose the mixtures ate used for the preservation of perishable products such, as cosmetics, pharmaceutical products and foods ([0034]).

Schmaus et al. differs from the instant claims insofar as they do not disclose wherein the 1,2-alkanediols are prepared from a bio-1-alkene having 7 to 20 carbon atoms.

Swern et al. disclose a process for synthesizing 1,2-oxtanedioll, 1,2-decandiol, 1,2-dodecandiol, 1,2-tetradecanediol, 1,2-hexadecanediol and 1,2-octadecanediol, from their corresponding 1-alkenes (p. 1504, column 2).

It would have been obvious to one of ordinary skill in the art before the effective filling date of the claimed invention to have used a known process of forming 1,2-alkane diols as the alkanediols of Schmaus since they are taught by Swern

Schmaus in view of Swern differs from the instant claims insofar as it does not disclose the 1,2-alkanediol as “bio”.

Wang et al. disclose due to the diminishing fossil fuel reservoirs together with the ever deteriorating environment associated with the worldwide fossil fuel based industry, it is imperative to make use of alternative feedstock for production of chemicals and fuels (p.758, Introduction).

It would have been obvious to one of ordinary skill in the art before the effective filing date of the claimed invention to have used “bio” based 1,2-alkanediols in the formulation of Schmaus in view of Swern motivated by the desire to preserve the environment as taught by Wang.

Claim 42-55 and 72-80 stand rejected under 35 U.S.C. 103 as being unpatentable over Lutz (US Patent Pub. 2012/0282207, previously disclosed) in view of Wang et al. (Green Chem., 2012, 14, pp 758-765, previously disclosed).

Lutz discloses an antimicrobial compositions and a product comprising a preservative effective amount of the said composition (Abstract). Lutz discloses household; fabric care and pharmaceutical products ([0043] and claim 25). Lutz discloses the composition comprises caprylyl glycol, which meets the limitation of 1,2-octanediol, pentylene glycol, hexylene glycol, and any combination thereof (reference claim 7). Lutz discloses the composition further comprises an antimicrobial mixture (reference claim 1).

Swern et al. disclose a process for synthesizing 1,2-oxtanedioll, 1,2-decandiol, 1,2-dodecandiol, 1,2-tetradecanediol, 1,2-hexadecanediol and 1,2-octadecanediol, from their corresponding 1-alkenes (p. 1504, column 2).

It would have been obvious to one of ordinary skill in the art before the effective filling date of the claimed invention to have used a known process of forming 1,2-alkane diols as the alkanediols of Lutz since they are taught by Swern

Lutz in view of Swern differs from the instant claims insofar as it does not disclose the 1,2-alkanediol as “bio”.

Wang et al. disclose due to the diminishing fossil fuel reservoirs together with the ever-deteriorating environment associated with the worldwide fossil-fuel-based industry, it is imperative to make use of alternative feedstock for the production of chemicals and fuels (p. 758, Introduction).

It would have been obvious to one of ordinary skill in the art before the effective filing date of the claimed invention to have used "bio" based 1,2-alkanediols in the formulation of Lutz in view of Swern motivated by the desire to preserve the environment as taught by Wang.

Conclusion

Any inquiry concerning this communication or earlier communications from the examiner should be directed to NANNETTE HOLLOMAN whose telephone number is (571)270-5231. The examiner can normally be reached Monday-Friday 9am-6pm.

Examiner interviews are available via telephone, in-person, and video conferencing using a USPTO supplied web-based collaboration tool. To schedule an interview, applicant is encouraged to use the USPTO Automated Interview Request (AIR) at http://www.uspto.gov/interviewpractice.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Frederick Krass can be reached on (571)272-0580. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of published or unpublished applications may be obtained from Patent Center. Unpublished application information in Patent Center is available to registered users. To file and manage patent submissions in Patent Center, visit: https://patentcenter.uspto.gov. Visit https://www.uspto.gov/patents/apply/patent-center for more information about Patent Center and https://www.uspto.gov/patents/docx for information about filing in DOCX format. For additional questions, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

/NANNETTE HOLLOMAN/
Primary Examiner, Art Unit 1612

Notice of References Cited	Application/Control No. 16/262,902	Applicant(s)/Patent Under Reexamination Burgo et al.	
	Examiner NANNETTE HOLLOMAN	Art Unit 1612	Page 1 of 1

U.S. PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Name	CPC Classification	US Classification
	A					
	B					
	C					
	D					
	E					
	F					
	G					
	H					
	I					
	J					
	K					
	L					
	M					

FOREIGN PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Country	Name	CPC Classification
	N					
	O					
	P					
	Q					
	R					
	S					
	T					

NON-PATENT DOCUMENTS

*		Include as applicable: Author, Title Date, Publisher, Edition or Volume, Pertinent Pages)
	U	Swern et al., Hydroxylation and epoxidation of some 1-olefins with per-acids, 1945, Journal of the American Chemical Society, vol. 67, pp. 1504-1507 (Year: 1945)
	V	
	W	
	X	

*A copy of this reference is not being furnished with this Office action. (See MPEP § 707.05(a).)
Dates in MM-YYYY format are publication dates. Classifications may be US or foreign.

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

CPC - Searched*		
Symbol	**Date**	**Examiner**
A01N	04/10/2021	NH
C07C	04/10/2021	NH
A61K	04/10/2021	NH
A61Q	04/10/2021	NH

CPC Combination Sets - Searched*		
Symbol	**Date**	**Examiner**

US Classification - Searched*			
Class	**Subclass**	**Date**	**Examiner**

* See search history printout included with this form or the SEARCH NOTES box below to determine the scope of the search.

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

Search Notes		
Search Notes	**Date**	**Examiner**
PALM Inventor Search	09/13/2020	NH
EAST Search	09/13/2020	NH
NPL Search	09/13/2020	NH
PALM Inventor Search	08/26/2021	NH
EAST Search	08/26/2021	NH
NPL Search	08/26/2021	NH
PALM Inventor Search	03/25/2022	NH
EAST Search	03/25/2022	NH
NPL Search	03/25/2022	NH

Interference Search			
US Class/CPC Symbol	**US Subclass/CPC Group**	**Date**	**Examiner**
C07C	11/18, 2527/054, 2527/167, 2527/173, 2531/02, 65/21,	04/10/2021	NH
A61K	8/345, 2300/00, 2800/31, 8/60, 2800/412, 8/11, 8/732, 8/738, 31/197, 8/44, 8/73, 2800/48, 8/25, 8/34, 8/585, 8/92, 8/9789, 9/014, 2800/30, 31/198, 36/28, 36/534, 36/67	04/10/2021	NH
A61Q	15/00, 19/00, 13/00, 11/00, 5/00, 19/007, 19/08, 19/02, 17/005, 17/04, 19/10, 5/02, 7/00	04/10/2021	NH

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Doc code: IDS
Doc description: Information Disclosure Statement (IDS) Filed

PTO/SB/08a (01-22)
Approved for use through 05/31/2024. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Not for submission under 37 CFR 1.99)

Application Number	16262902
Filing Date	2019-01-30
First Named Inventor	BURGO, Rocco V.
Art Unit	1612
Examiner Name	HOLLOMAN, Nannette
Attorney Docket Number	INLX.0198U2/484799

U.S.PATENTS Remove

Examiner Initial*	Cite No	Patent Number	Kind Code[1]	Issue Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear
	1					

If you wish to add additional U.S. Patent citation information please click the Add button. Add

U.S.PATENT APPLICATION PUBLICATIONS Remove

Examiner Initial*	Cite No	Publication Number	Kind Code[1]	Publication Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear
	1	20090143489	A1	2009-06-04	WINN, Daniel	
	2	20090004122	A1	2009-01-01	MODAK, Shanta, et al.	
	3	20120238788	A1	2012-09-20	WRIGHT, Michael E.	

If you wish to add additional U.S. Published Application citation information please click the Add button. Add

FOREIGN PATENT DOCUMENTS Remove

Examiner Initial*	Cite No	Foreign Document Number[3]	Country Code[2] i	Kind Code[4]	Publication Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear	T[5]
	1	3045161	EP	A1	2016-07-20	SYMRISE AG		

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Not for submission under 37 CFR 1.99)

Application Number	16262902
Filing Date	2019-01-30
First Named Inventor	BURGO, Rocco V.
Art Unit	1612
Examiner Name	HOLLOMAN, Nannette
Attorney Docket Number	INLX.0198U2/484799

If you wish to add additional Foreign Patent Document citation information please click the Add button [Add]

NON-PATENT LITERATURE DOCUMENTS [Remove]

Examiner Initials*	Cite No	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc), date, pages(s), volume-issue number(s), publisher, city and/or country where published.	T[5]
	1	Supplementary European Search Report from European Application No. EP 19 74 7575; mailing date of February 24, 2022; Paul Campbell (Examiner)	

If you wish to add additional non-patent literature document citation information please click the Add button [Add]

EXAMINER SIGNATURE

Examiner Signature	/NANNETTE HOLLOMAN/	Date Considered	03/25/2022

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through a citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

[1] See Kind Codes of USPTO Patent Documents at www.USPTO.GOV or MPEP 901.04. [2] Enter office that issued the document, by the two-letter code (WIPO Standard ST.3). [3] For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. [4] Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. [5] Applicant is to place a check mark here if English language translation is attached.

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Not for submission under 37 CFR 1.99)

Application Number	16262902
Filing Date	2019-01-30
First Named Inventor	BURGO, Rocco V.
Art Unit	1612
Examiner Name	HOLLOMAN, Nannette
Attorney Docket Number	INLX.0198U2/484799

CERTIFICATION STATEMENT

Please see 37 CFR 1.97 and 1.98 to make the appropriate selection(s):

☒ That each item of information contained in the information disclosure statement was first cited in any communication from a foreign patent office in a counterpart foreign application not more than three months prior to the filing of the information disclosure statement. See 37 CFR 1.97(e)(1).

OR

☐ That no item of information contained in the information disclosure statement was cited in a communication from a foreign patent office in a counterpart foreign application, and, to the knowledge of the person signing the certification after making reasonable inquiry, no item of information contained in the information disclosure statement was known to any individual designated in 37 CFR 1.56(c) more than three months prior to the filing of the information disclosure statement. See 37 CFR 1.97(e)(2).

See attached certification statement.

The fee set forth in 37 CFR 1.17 (p) has been submitted herewith.

A certification statement is not submitted herewith.

SIGNATURE

A signature of the applicant or representative is required in accordance with CFR 1.33, 10.18. Please see CFR 1.4(d) for the form of the signature.

Signature	/kathryn a warner/	Date (YYYY-MM-DD)	2022-03-16
Name/Print	Kathryn A. Warner	Registration Number	59,858

This collection of information is required by 37 CFR 1.97 and 1.98. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 1 hour to complete, including gathering, preparing and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

Privacy Act Statement

The Privacy Act of 1974 (P.L. 93-579) requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C. 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether the Freedom of Information Act requires disclosure of these record s.

2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.

3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.

4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).

5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.

6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).

7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.

8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspections or an issued patent.

9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

Web Search History

date, time	web site	search string
3/25/2022 1:14:49 PM	Google	hydroxylation and epoxidation of some 1-olefins with per acids
3/25/2022 2:22:17 PM	Google Scholar	bio alkanediol

EAST Search History

EAST Search History (Prior Art)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
L1	993	((("BURGO") near3 ("Rocco")) OR (("WRIGHT") near3 ("Michael")) OR (("MOSSER") near3 ("Gary")) OR (("FEVOLA") near3 ("Michael"))).INV.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	OFF	2022/03/25 14:21
L2	6	l1 and alkanediol	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2022/03/25 14:21
S1	937	((("BURGO") near3 ("Rocco")) OR (("WRIGHT") near3 ("Michael")) OR (("MOSSER") near3 ("Gary")) OR (("FEVOLA") near3 ("Michael"))).INV.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	OFF	2020/07/07 19:12
S2	378	(bio with alkene) bioalkene	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/07/07 19:19
S3	6,577	regioselectivity	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/09 14:30
S4	13	regioselectivity same alkanediol	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/09 14:30
S5	7	regioselectivity same (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/09 14:47
S6	1	("7385092").PN.	USPAT; USOCR	OR	OFF	2020/09/09 14:53
S7	2	wo-2016007196-$.did.	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	OFF	2020/09/11 11:19

S8	6	alkanediol same (household and pharmaceutical and cosmetic)	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:25
S9	3	(household and pharmaceutical and cosmetic) same (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26
S10	336	(household and pharmaceutical and cosmetic) and (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26
S11	6	((household and pharmaceutical and cosmetic) and (octanediol or (caprylyl adj glycol))).clm.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:26
S12	11	((household and pharmaceutical and cosmetic) and (octanediol or ((caprylyl or hexylene or decylene) adj glycol) or alkanediol)).clm.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2020/09/11 17:47
S13	946	((("BURGO") near3 ("Rocco")) OR (("WRIGHT") near3 ("Michael")) OR (("MOSSER") near3 ("Gary")) OR (("FEVOLA") near3 ("Michael"))).INV.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	OFF	2020/09/11 19:14
S14	3	S13 and alkanediol	US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT	OR	ON	2020/09/11 19:18
S15	7	(("8912373") or ("9856461") or ("4234752") or ("20050065389") or ("20030065233") or ("20060161025") or ("20140066666")).PN.	US-PGPUB; USPAT; USOCR	OR	OFF	2020/09/13 13:37
S16	42,261	decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/02/27 23:31
S17	13,209	S16 and (preservative or pharmaceutical or cosmetic)	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/02/27 23:32

S18	1,488	S17 and "bio"	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/02/27 23:32
S19	67	"bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol)))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/04/10 19:56
S20	549,665	((A01N47/44 OR A01N37/36 OR A01N59/16 OR A01N31/02 OR A01N25/34 OR A01N59/00 OR A01N61/00 OR A01N25/00 OR A01N2300/00 OR A01N25/10 OR C07C31/20 OR C07C1/24 OR C07C11/02 OR C07C29/48 OR C07C29/80 OR C07C263/12 OR C07C265/14 OR C07C2521/04 OR C07C11/107 OR C07C29/04 OR C07C247/24 OR C07C11/167 OR C07C11/18 OR C07C2527/054 OR C07C2527/167 OR C07C2527/173 OR C07C2531/02 OR C07C65/21 OR A61K8/345 OR A61K2300/00 OR A61K2800/31 OR A61K8/60 OR A61K2800/412 OR A61K8/11 OR A61K8/732 OR A61K8/738 OR A61K31/197 OR A61K8/44 OR A61K8/73 OR A61K2800/48 OR A61K8/25 OR A61K8/34 OR A61K8/585 OR A61K8/92 OR A61K8/9789 OR A61K9/0014 OR A61K2800/30 OR A61K31/198 OR A61K36/28 OR A61K36/534 OR A61K36/67 OR A61K36/886 OR A61K45/06 OR A61K47/10 OR A61K8/24 OR A61K8/362 OR A61K8/37 OR A61K8/41 OR A61K8/42 OR A61K8/4973 OR A61K8/498 OR A61K8/891 OR A61K9/06 OR A61K2800/10 OR A61K2800/28 OR A61K2800/524 OR A61K2800/56 OR A61K2800/87 OR A61K2800/874 OR A61K31/327 OR A61K47/22 OR A61K47/44 OR A61K47/46 OR A61K8/0216 OR A61K8/046 OR A61K8/19 OR A61K8/20 OR A61K8/23 OR A61K8/26 OR A61K8/31 OR A61K8/64 OR A61K8/678 OR A61K8/735 OR A61K8/8111 OR A61K8/8147 OR A61K8/893 OR A61K8/895 OR A61K8/922 OR A61K8/9794 OR A61K9/107 OR A61K2035/115 OR	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/04/10 19:57

		A61K2800/43 OR A61K2800/522 OR A61K2800/592 OR A61K2800/5922 OR A61K2800/594 OR A61K2800/596 OR A61K2800/624 OR A61K2800/805 OR A61K31/192 OR A61K31/355 OR A61K31/60 OR A61K33/30 OR A61K35/745 OR A61K35/747 OR A61K36/185 OR A61K36/53 OR A61K47/02 OR A61K47/06 OR A61K47/12 OR A61K47/14 OR A61K47/183 OR A61K47/24 OR A61K47/34 OR A61K47/38 OR A61K49/0006 OR A61K49/0008 OR A61K8/06 OR A61K8/062 OR A61K8/28 OR A61K8/29 OR A61K8/342 OR A61K8/347 OR A61K8/365 OR A61K8/368 OR A61K8/375 OR A61K8/416 OR A61K8/445 OR A61K8/463 OR A61K8/49 OR A61K8/4926 OR A61K8/63 OR A61K8/66 OR A61K8/675 OR A61K8/8152 OR A61K8/8194 OR A61K8/927 OR A61K8/965 OR A61K8/97 OR A61K9/08 OR A61Q15/00 OR A61Q19/00 OR A61Q13/00 OR A61Q11/00 OR A61Q5/00 OR A61Q19/007 OR A61Q19/08 OR A61Q19/02 OR A61Q17/005 OR A61Q17/04 OR A61Q19/10 OR A61Q5/02 OR A61Q7/00).CPC.)				
S21	42	S20 and ("bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))))	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2021/04/10 19:57
S24	2	(("9242226") or ("20050222276")).PN.	US-PGPUB; USPAT; USOCR	OR	OFF	2021/08/14 21:51
S25	4	("20050069997" | "7385092" | "8450543" | "9732295").PN.	US-PGPUB; USPAT	OR	OFF	2021/11/26 11:34
S26	4	("20050069997" | "7385092" | "8450543" | "9732295").PN.	US-PGPUB; USPAT	OR	OFF	2021/11/26 11:35
S27	48,937	"bio".ti.	US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT	OR	ON	2022/01/19 12:50
S28	43	(natural or (natural adj derived)) with "1,2-alkanediol"	US-PGPUB; USPAT; FPRS; EPO;	OR	ON	2022/01/19 12:51

			JPO; DERWENT			
S29	2	(("8912373") or ("9242226")).PN.	USPAT; USOCR	OR	OFF	2022/01/19 12:59
S30	3	("20090004122" | "20090143489" | "20120238788").PN.	US-PGPUB; USPAT	OR	OFF	2022/03/23 20:39

EAST Search History (Interference)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
S22	125,400	((A01N47/44 OR A01N37/36 OR A01N59/16 OR A01N31/02 OR A01N25/34 OR A01N59/00 OR A01N61/00 OR A01N25/00 OR A01N2300/00 OR A01N25/10 OR C07C31/20 OR C07C1/24 OR C07C11/02 OR C07C29/48 OR C07C29/80 OR C07C263/12 OR C07C265/14 OR C07C2521/04 OR C07C11/107 OR C07C29/04 OR C07C247/24 OR C07C11/167 OR C07C11/18 OR C07C2527/054 OR C07C2527/167 OR C07C2527/173 OR C07C2531/02 OR C07C65/21 OR A61K8/345 OR A61K2300/00 OR A61K2800/31 OR A61K8/60 OR A61K2800/412 OR A61K8/11 OR A61K8/732 OR A61K8/738 OR A61K31/197 OR A61K8/44 OR A61K8/73 OR A61K2800/48 OR A61K8/25 OR A61K8/34 OR A61K8/585 OR A61K8/92 OR A61K8/9789 OR A61K9/0014 OR A61K2800/30 OR A61K31/198 OR A61K36/28 OR A61K36/534 OR A61K36/67 OR A61K36/886 OR A61K45/06 OR A61K47/10 OR A61K8/24 OR A61K8/362 OR A61K8/37 OR A61K8/41 OR A61K8/42 OR A61K8/4973 OR A61K8/498 OR A61K8/891 OR A61K9/06 OR A61K2800/10 OR A61K2800/28 OR A61K2800/524 OR A61K2800/56 OR A61K2800/87 OR A61K2800/874 OR A61K31/327 OR A61K47/22 OR A61K47/44 OR A61K47/46 OR A61K8/0216 OR A61K8/046 OR A61K8/19 OR A61K8/20 OR A61K8/23 OR A61K8/26 OR A61K8/31 OR A61K8/64 OR A61K8/678 OR A61K8/735 OR A61K8/8111 OR A61K8/8147 OR A61K8/893 OR A61K8/895 OR A61K8/922 OR A61K8/9794 OR A61K9/107 OR A61K2035/115 OR A61K2800/43 OR A61K2800/522 OR A61K2800/592 OR A61K2800/5922 OR A61K2800/594 OR A61K2800/596 OR A61K2800/624 OR A61K2800/805 OR A61K31/192 OR	USPAT	OR	ON	2021/04/10 19:59

		A61K31/355 OR A61K31/60 OR A61K33/30 OR A61K35/745 OR A61K35/747 OR A61K36/185 OR A61K36/53 OR A61K47/02 OR A61K47/06 OR A61K47/12 OR A61K47/14 OR A61K47/183 OR A61K47/24 OR A61K47/34 OR A61K47/38 OR A61K49/0006 OR A61K49/0008 OR A61K8/06 OR A61K8/062 OR A61K8/28 OR A61K8/29 OR A61K8/342 OR A61K8/347 OR A61K8/365 OR A61K8/368 OR A61K8/375 OR A61K8/416 OR A61K8/445 OR A61K8/463 OR A61K8/49 OR A61K8/4926 OR A61K8/63 OR A61K8/66 OR A61K8/675 OR A61K8/8152 OR A61K8/8194 OR A61K8/927 OR A61K8/965 OR A61K8/97 OR A61K9/08 OR A61Q15/00 OR A61Q19/00 OR A61Q13/00 OR A61Q11/00 OR A61Q5/00 OR A61Q19/007 OR A61Q19/08 OR A61Q19/02 OR A61Q17/005 OR A61Q17/04 OR A61Q19/10 OR A61Q5/02 OR A61Q7/00).CPC.)				
S23	10	S22 and ("bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))))	USPAT	OR	ON	2021/04/10 19:59

3/25/2022 2:22:53 PM
C:\Users\nholloman\Documents\EAST\Workspaces\16262902.wsp

PTO/SB/06 (09-11)
Approved for use through 1/31/2014. OMB 0651-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

PATENT APPLICATION FEE DETERMINATION RECORD Substitute for Form PTO-875	Application or Docket Number 16/262,902	Filing Date 01/30/2019	☐ To be Mailed

ENTITY: ☑ LARGE ☐ SMALL ☐ MICRO

APPLICATION AS FILED - PART I

FOR	(Column 1) NUMBER FILED	(Column 2) NUMBER EXTRA		RATE ($)	FEE ($)
☐ BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A		N/A	
☐ SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A		N/A	
☐ EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A		N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	minus 20 =	*		x $100 =	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	minus 3 =	*		x $460 =	
☐ APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $310 ($155 for small entity) for each additional 50 sheets or fraction thereof. See 35 U.S.C. 41(a)(1)(G) and 37 CFR 1.16(s).				
☐ MULTIPLE DEPENDENT CLAIM PRESENT (37 CFR 1.16(j))					
* If the difference in column 1 is less than zero, enter "0" in column 2.				TOTAL	

APPLICATION AS AMENDED - PART II

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT	06/03/2022	CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	* 24	Minus	** 24	= 0		x $100 =	0
	Independent (37 CFR 1.16(h))	* 1	Minus	*** 3	= 0		x $480 =	0
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	0

		(Column 1)		(Column 2)	(Column 3)			
AMENDMENT		CLAIMS REMAINING AFTER AMENDMENT		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA		RATE ($)	ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	*	Minus	**	=		x $0 =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=		x $0 =	
	☐ Application Size Fee (37 CFR 1.16(s))							
	☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							
							TOTAL ADD'L FEE	

* If the entry in column 1 is less than the entry in column 2, write "0" in column 3.	LIE
** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 20, enter "20".	/CAROLYN E THOMAS/
*** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 3, enter "3".	
The "Highest Number Previously Paid For" (Total or Independent) is the highest number found in the appropriate box in column 1.	

This collection of information is required by 37 CFR 1.16. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

First Named Inventor: **Rocco V. Burgo**	Attorney Docket No: **INLX.0198U2/00484799**
Application No.: **16/262,902**	Confirmation No.: **1028**
Filing Date: **January 30, 2019**	Art Unit: **1612**
Title: ***Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same***	Examiner: **HOLLOMAN, Nannette**

VIA EFS

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

RESPONSE TO OFFICE ACTION UNDER 37 C.F.R. § 1.111

In response to the Office Action mailed March 30, 2022, Applicants respectfully request reconsideration of this application in view of the following claim amendments and remarks:

A marked-up **Listing of the Claims** begin on page 2.

Remarks begin on page 5.

LISTING OF THE CLAIMS

This listing of claims will replace all prior versions, and listings, of claims in the application.

What is Claimed is:

1-87. (Canceled)

88. (Previously presented) A preservative composition, comprising about 60% to about 99% of a bio-1,2-alkanediol having a carbon chain length of 7 to 20 carbon atoms, wherein the carbon atoms of the bio-1,2-alkanediol are derived from a plant source and are not petrochemically derived, and wherein the preservative composition comprises less than 20% of a corresponding bio-2,3-alkanediol having the same carbon chain length as the bio-1,2-alkanediol.

89. (Previously presented) The preservative composition of claim 88, wherein the bio-1,2-alkanediol has a carbon chain length of 7 to 14 carbon atoms.

90. (Previously presented) The preservative composition of claim 89, comprising less than about 8% of the corresponding bio-2,3-alkanediol having the same carbon chain length as the bio-1,2-alkanediol.

91. (Previously presented) The preservative composition of claim 90, comprising less than about 5% of the corresponding bio-2,3-alkanediol having the same carbon chain length as the bio-1,2-alkanediol.

92. (New) The preservative composition of claim 89, wherein the bio-1,2-alkanediol is selected from the group consisting of: bio-1,2-heptanediol, bio-1,2-octanediol, bio-1,2-decanediol, bio-1,2-dodecanediol, and combinations thereof.

93. (New) The preservative composition of claim 92, wherein the bio-1,2-alkanediol is bio-1,2-heptanediol.

94. (New) The preservative composition of claim 93, comprising less than about 5% of bio-2,3-heptanediol.

95. (New) The preservative composition of claim 94, further comprising an antimicrobial compound different from the bio-1,2-heptanediol.

96. (New) The preservative composition of claim 94, further comprising a bio-compound selected from the group consisting of: a bio-organic acid, bio-hydroxamic acid, a bio-alkylhydroxamic acid, a bio-terpenoid compound having 5 to 20 carbon atoms, bio-1,3-

propanediol, bio-1,2-butanediol, bio-1,2-pentanediol, bio-1,2-hexanediol, and combinations thereof.

97. (New) The preservative composition of claim 96, comprising about 75% to about 99% of the bio-1,2-heptanediol.

98. (New) The preservative composition of claim 92, wherein the bio-1,2-alkanediol is bio-1,2-octanediol.

99. (New) The preservative composition of claim 98, comprising less than about 5% of bio-2,3-octanediol.

100. (New) The preservative composition of claim 99, further comprising an antimicrobial compound different from the bio-1,2-octanediol.

101. (New) The preservative composition of claim 100, further comprising a bio-compound selected from the group consisting of: a bio-organic acid, bio-hydroxamic acid, a bio-alkylhydroxamic acid, a bio-terpenoid compound having 5 to 20 carbon atoms, bio-1,3-propanediol, bio-1,2-butanediol, bio-1,2-pentanediol, bio-1,2-hexanediol, and combinations thereof.

102. (New) The preservative composition of claim 101, comprising about 75% to about 99% of the bio-1,2-octanediol.

103. (New) The preservative composition of claim 91, further comprising an antimicrobial compound different from the bio-1,2-alkanediol.

104. (New) The preservative composition of claim 103, further comprising a bio-compound selected from the group consisting of: a bio-organic acid, bio-hydroxamic acid, a bio-alkylhydroxamic acid, a bio-terpenoid compound having 5 to 20 carbon atoms, bio-1,3-propanediol, bio-1,2-butanediol, bio-1,2-pentanediol, bio-1,2-hexanediol, and combinations thereof.

105. (New) The preservative composition of claim 104, comprising about 75% to about 99% of the bio-1,2-alkanediol.

106. (New) A personal care composition comprising the preservative composition of claim 88 in a concentration of about 0.1% to about 10%.

107. (New) A personal care composition comprising the preservative composition of claim 94 in a concentration of about 0.1% to about 10%.

108. (New) A personal care composition comprising the preservative composition of claim 97 in a concentration of about 0.1% to about 10%.

109. (New) A personal care composition comprising the preservative composition of claim 99 in a concentration of about 0.1% to about 10%.

110. (New) A personal care composition comprising the preservative composition of claim 102 in a concentration of about 0.1% to about 10%.

111. (New) A personal care composition comprising the preservative composition of claim 105 in a concentration of about 0.1% to about 10%.

REMARKS

Application and Claims Status

Upon entry of the foregoing claim amendments, claims 88-111 are pending. Claims 1-87 have been cancelled.

Written support for new claims 92-111 can be found in the as-filed specification. Specifically, the Examiner's attention is drawn to the following non-limiting exemplary paragraphs of the as-filed specification:

- written support for new claims 92, 93, and 98 can be found, *inter alia*, at ¶¶ [0029]-[0031], [0047]-[0048], [0066]-[0068], [0121], and [0129];
- written support for new claims 94 and 99 can be found, *inter alia*, at ¶¶ [0033], [0047]-[0049], [0059], [0066], [0092], [0129], [0131], Table 3, and Fig. 5;
- written support for new claims 95, 96, 100, 101, 103, and 104 can be found, *inter alia*, at ¶¶ [0047]-[0049], [0051], and [0094];
- written support for new claims 97, 102, and 105 can be found, *inter alia*, at ¶¶ [0059], [0090], [0129], [0131], Table 3, and Fig. 5; and
- written support for new claims 106-111 can be found, *inter alia*, at ¶¶ [0095], [0100], [0119], of the as-filed specification.

Claims 42-55 and 72-80, which were rejected as unpatentable under 35 U.S.C. § 103, have been cancelled. Applicants note that the cancellation of claims 42-55 and 72-80 is being made solely to advance the prosecution of the instant application and does not constitute any waiver or surrender of any subject matter that Applicants may be entitled to pursue, at their option, in a future submission such as a continuing or reissue application. The cancellation of claims 42-55 and 72-80 similarly does not constitute any agreement or acquiescence as to the merits of the Office Action. Accordingly, Applicants expressly reserve the right to pursue the scope of any of the original, previously presented, withdrawn, or cancelled claims, as well as other claim scope, in a future submission.

In the Office Action dated March 30, 2022, the Office noted that "[c]laims 88-91 appear to be free from art." Office Action at 2. Because new claims 92-111 depend from claims 88 and 89, Applicants respectfully submit that these claims should be similarly free from the prior art. Thus, Applicants respectfully submit that new claims 88-111 are in condition for allowance.

CONCLUSION

In view of the foregoing claim amendments and remarks, Applicants respectfully submit that the pending claims are in condition for allowance and Notice of Allowance is earnestly solicited. The Examiner is invited to contact the undersigned attorney with any questions, comments or suggestions relating to the referenced patent application.

This response is being submitted on June 3, 2022. In the event the Commissioner determines that any additional fee(s) are required for any reason, the Commissioner is hereby authorized to deduct said fee(s) from Deposit Account No.: 50-3111/Matter No.: 00484799.

Respectfully submitted,

Date: June 3, 2022

/Aaron Lukas, Reg.# 59,443/

Aaron S. Lukas, Ph.D.
Reg. No. 59,443
COZEN O'CONNOR
1200 Nineteenth Street N.W.
Washington, D.C. 20036
Tel: (202) 912-4823
Fax: (202) 861-1905

Electronic Patent Application Fee Transmittal

Application Number:	16262902
Filing Date:	30-Jan-2019
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Filer:	Aaron Scott Lukas/Valerie Foster
Attorney Docket Number:	INLX.0198U2/00484799

Filed as Large Entity

Filing Fees for Utility under 35 USC 111(a)

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Pages:				
Claims:				
CLAIMS IN EXCESS OF 20	1202	1	100	100
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Extension-of-Time:				
Miscellaneous:				
			Total in USD ($)	**100**

Electronic Acknowledgement Receipt

EFS ID:	45871537
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	171052
Filer:	Aaron Scott Lukas/Valerie Foster
Filer Authorized By:	Aaron Scott Lukas
Attorney Docket Number:	INLX.0198U2/00484799
Receipt Date:	03-JUN-2022
Filing Date:	30-JAN-2019
Time Stamp:	15:04:19
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	CARD
Payment was successfully received in RAM	$100
RAM confirmation Number	E202263F04480642
Deposit Account	
Authorized User	

The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows:

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1		INLX_0198U2_00484799_ResponsetoOfficeAction.pdf	210571 48caf7da6d6f6f0b04c6d5fd56082e2e2350128e	yes	6

	Multipart Description/PDF files in .zip description		
	Document Description	**Start**	**End**
	Amendment/Request for Reconsideration-After Non-Final Rejection	1	1
	Claims	2	4
	Applicant Arguments/Remarks Made in an Amendment	5	6

Warnings:					
Information:					
2	Fee Worksheet (SB06)	fee-info.pdf	38132 e06f2b335dbecfde7bb2ca9e53822c5201b0e13c	no	2
Warnings:					
Information:					
			Total Files Size (in bytes):	248703	

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

NOTICE OF ALLOWANCE AND FEE(S) DUE

171052 7590 06/23/2022

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

EXAMINER
HOLLOMAN, NANNETTE

ART UNIT	PAPER NUMBER
1612	

DATE MAILED: 06/23/2022

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	INLX.0198U2/00484799	1028

TITLE OF INVENTION: Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same

APPLN. TYPE	ENTITY STATUS	ISSUE FEE DUE	PUBLICATION FEE DUE	PREV. PAID ISSUE FEE	TOTAL FEE(S) DUE	DATE DUE
nonprovisional	UNDISCOUNTED	$1200	$0.00	$0.00	$1200	09/23/2022

THE APPLICATION IDENTIFIED ABOVE HAS BEEN EXAMINED AND IS ALLOWED FOR ISSUANCE AS A PATENT. PROSECUTION ON THE MERITS IS CLOSED. THIS NOTICE OF ALLOWANCE IS NOT A GRANT OF PATENT RIGHTS. THIS APPLICATION IS SUBJECT TO WITHDRAWAL FROM ISSUE AT THE INITIATIVE OF THE OFFICE OR UPON PETITION BY THE APPLICANT. SEE 37 CFR 1.313 AND MPEP 1308.

THE ISSUE FEE AND PUBLICATION FEE (IF REQUIRED) MUST BE PAID WITHIN THREE MONTHS FROM THE MAILING DATE OF THIS NOTICE OR THIS APPLICATION SHALL BE REGARDED AS ABANDONED. THIS STATUTORY PERIOD CANNOT BE EXTENDED. SEE 35 U.S.C. 151. THE ISSUE FEE DUE INDICATED ABOVE DOES NOT REFLECT A CREDIT FOR ANY PREVIOUSLY PAID ISSUE FEE IN THIS APPLICATION. IF AN ISSUE FEE HAS PREVIOUSLY BEEN PAID IN THIS APPLICATION (AS SHOWN ABOVE), THE RETURN OF PART B OF THIS FORM WILL BE CONSIDERED A REQUEST TO REAPPLY THE PREVIOUSLY PAID ISSUE FEE TOWARD THE ISSUE FEE NOW DUE.

HOW TO REPLY TO THIS NOTICE:

I. Review the ENTITY STATUS shown above. If the ENTITY STATUS is shown as SMALL or MICRO, verify whether entitlement to that entity status still applies.

If the ENTITY STATUS is the same as shown above, pay the TOTAL FEE(S) DUE shown above.

If the ENTITY STATUS is changed from that shown above, on PART B - FEE(S) TRANSMITTAL, complete section number 5 titled "Change in Entity Status (from status indicated above)".

For purposes of this notice, small entity fees are 1/2 the amount of undiscounted fees, and micro entity fees are 1/2 the amount of small entity fees.

II. PART B - FEE(S) TRANSMITTAL, or its equivalent, must be completed and returned to the United States Patent and Trademark Office (USPTO) with your ISSUE FEE and PUBLICATION FEE (if required). If you are charging the fee(s) to your deposit account, section "4b" of Part B - Fee(s) Transmittal should be completed. If an equivalent of Part B is filed, a request to reapply a previously paid issue fee must be clearly made, and delays in processing may occur due to the difficulty in recognizing the paper as an equivalent of Part B.

III. All communications regarding this application must give the application number. Please direct all communications prior to issuance to Mail Stop ISSUE FEE unless advised to the contrary.

IMPORTANT REMINDER: Maintenance fees are due in utility patents issuing on applications filed on or after Dec. 12, 1980. It is patentee's responsibility to ensure timely payment of maintenance fees when due. More information is available at www.uspto.gov/PatentMaintenanceFees.

PART B - FEE(S) TRANSMITTAL

Complete and send this form, together with applicable fee(s), by mail or fax, or via EFS-Web.

By mail, send to: Mail Stop ISSUE FEE
Commissioner for Patents
P.O. Box 1450
Alexandria, Virginia 22313-1450

By fax, send to: (571)-273-2885

INSTRUCTIONS: This form should be used for transmitting the ISSUE FEE and PUBLICATION FEE (if required). Blocks 1 through 5 should be completed where appropriate. All further correspondence including the Patent, advance orders and notification of maintenance fees will be mailed to the current correspondence address as indicated unless corrected below or directed otherwise in Block 1, by (a) specifying a new correspondence address; and/or (b) indicating a separate "FEE ADDRESS" for maintenance fee notifications.

CURRENT CORRESPONDENCE ADDRESS (Note: Use Block 1 for any change of address)

171052 7590 06/23/2022

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

Note: A certificate of mailing can only be used for domestic mailings of the Fee(s) Transmittal. This certificate cannot be used for any other accompanying papers. Each additional paper, such as an assignment or formal drawing, must have its own certificate of mailing or transmission.

Certificate of Mailing or Transmission

I hereby certify that this Fee(s) Transmittal is being deposited with the United States Postal Service with sufficient postage for first class mail in an envelope addressed to the Mail Stop ISSUE FEE address above, or being transmitted to the USPTO via EFS-Web or by facsimile to (571) 273-2885, on the date below.

(Typed or printed name)
(Signature)
(Date)

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	INLX.0198U2/00484799	1028

TITLE OF INVENTION: Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same

APPLN. TYPE	ENTITY STATUS	ISSUE FEE DUE	PUBLICATION FEE DUE	PREV. PAID ISSUE FEE	TOTAL FEE(S) DUE	DATE DUE
nonprovisional	UNDISCOUNTED	$1200	$0.00	$0.00	$1200	09/23/2022

EXAMINER	ART UNIT	CLASS-SUBCLASS
HOLLOMAN, NANNETTE	1612	424-049000

1. Change of correspondence address or indication of "Fee Address" (37 CFR 1.363).

❑ Change of correspondence address (or Change of Correspondence Address form PTO/AIA/122 or PTO/SB/122) attached.

❑ "Fee Address" indication (or "Fee Address" Indication form PTO/AIA/47 or PTO/SB/47; Rev 03-02 or more recent) attached. **Use of a Customer Number is required.**

2. For printing on the patent front page, list
(1) The names of up to 3 registered patent attorneys or agents OR, alternatively,
(2) The name of a single firm (having as a member a registered attorney or agent) and the names of up to 2 registered patent attorneys or agents. If no name is listed, no name will be printed.

1 _____
2 _____
3 _____

3. ASSIGNEE NAME AND RESIDENCE DATA TO BE PRINTED ON THE PATENT (print or type)

PLEASE NOTE: Unless an assignee is identified below, no assignee data will appear on the patent. If an assignee is identified below, the document must have been previously recorded, or filed for recordation, as set forth in 37 CFR 3.11 and 37 CFR 3.81(a). Completion of this form is NOT a substitute for filing an assignment.

(A) NAME OF ASSIGNEE (B) RESIDENCE: (CITY and STATE OR COUNTRY)

Please check the appropriate assignee category or categories (will not be printed on the patent) : ❑ Individual ❑ Corporation or other private group entity ❑ Government

4a. Fees submitted: ❑ Issue Fee ❑ Publication Fee (if required) ❑ Advance Order - # of Copies _____

4b. Method of Payment: *(Please first reapply any previously paid fee shown above)*

❑ Electronic Payment via EFS-Web ❑ Enclosed check ❑ Non-electronic payment by credit card (Attach form PTO-2038)

❑ The Director is hereby authorized to charge the required fee(s), any deficiency, or credit any overpayment to Deposit Account No. _____

5. **Change in Entity Status** (from status indicated above)

❑ Applicant certifying micro entity status. See 37 CFR 1.29

❑ Applicant asserting small entity status. See 37 CFR 1.27

❑ Applicant changing to regular undiscounted fee status.

NOTE: Absent a valid certification of Micro Entity Status (see forms PTO/SB/15A and 15B), issue fee payment in the micro entity amount will not be accepted at the risk of application abandonment.
NOTE: If the application was previously under micro entity status, checking this box will be taken to be a notification of loss of entitlement to micro entity status.
NOTE: Checking this box will be taken to be a notification of loss of entitlement to small or micro entity status, as applicable.

NOTE: This form must be signed in accordance with 37 CFR 1.31 and 1.33. See 37 CFR 1.4 for signature requirements and certifications.

Authorized Signature _____ Date _____

Typed or printed name _____ Registration No. _____

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	INLX.0198U2/00484799	1028

171052 7590 06/23/2022

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

EXAMINER	
HOLLOMAN, NANNETTE	
ART UNIT	PAPER NUMBER
1612	

DATE MAILED: 06/23/2022

Determination of Patent Term Adjustment under 35 U.S.C. 154 (b)
(Applications filed on or after May 29, 2000)

The Office has discontinued providing a Patent Term Adjustment (PTA) calculation with the Notice of Allowance.

Section 1(h)(2) of the AIA Technical Corrections Act amended 35 U.S.C. 154(b)(3)(B)(i) to eliminate the requirement that the Office provide a patent term adjustment determination with the notice of allowance. See Revisions to Patent Term Adjustment, 78 Fed. Reg. 19416, 19417 (Apr. 1, 2013). Therefore, the Office is no longer providing an initial patent term adjustment determination with the notice of allowance. The Office will continue to provide a patent term adjustment determination with the Issue Notification Letter that is mailed to applicant approximately three weeks prior to the issue date of the patent, and will include the patent term adjustment on the patent. Any request for reconsideration of the patent term adjustment determination (or reinstatement of patent term adjustment) should follow the process outlined in 37 CFR 1.705.

Any questions regarding the Patent Term Extension or Adjustment determination should be directed to the Office of Patent Legal Administration at (571)-272-7702. Questions relating to issue and publication fee payments should be directed to the Customer Service Center of the Office of Patent Publication at 1-(888)-786-0101 or (571)-272-4200.

OMB Clearance and PRA Burden Statement for PTOL-85 Part B

The Paperwork Reduction Act (PRA) of 1995 requires Federal agencies to obtain Office of Management and Budget approval before requesting most types of information from the public. When OMB approves an agency request to collect information from the public, OMB (i) provides a valid OMB Control Number and expiration date for the agency to display on the instrument that will be used to collect the information and (ii) requires the agency to inform the public about the OMB Control Number's legal significance in accordance with 5 CFR 1320.5(b).

The information collected by PTOL-85 Part B is required by 37 CFR 1.311. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 30 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, Virginia 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, Virginia 22313-1450. Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

Privacy Act Statement

The Privacy Act of 1974 (P.L. 93-579) requires that you be given certain information in connection with your submission of the attached form related to a patent application or patent. Accordingly, pursuant to the requirements of the Act, please be advised that: (1) the general authority for the collection of this information is 35 U.S.C. 2(b)(2); (2) furnishing of the information solicited is voluntary; and (3) the principal purpose for which the information is used by the U.S. Patent and Trademark Office is to process and/or examine your submission related to a patent application or patent. If you do not furnish the requested information, the U.S. Patent and Trademark Office may not be able to process and/or examine your submission, which may result in termination of proceedings or abandonment of the application or expiration of the patent.

The information provided by you in this form will be subject to the following routine uses:

1. The information on this form will be treated confidentially to the extent allowed under the Freedom of Information Act (5 U.S.C. 552) and the Privacy Act (5 U.S.C 552a). Records from this system of records may be disclosed to the Department of Justice to determine whether disclosure of these records is required by the Freedom of Information Act.
2. A record from this system of records may be disclosed, as a routine use, in the course of presenting evidence to a court, magistrate, or administrative tribunal, including disclosures to opposing counsel in the course of settlement negotiations.
3. A record in this system of records may be disclosed, as a routine use, to a Member of Congress submitting a request involving an individual, to whom the record pertains, when the individual has requested assistance from the Member with respect to the subject matter of the record.
4. A record in this system of records may be disclosed, as a routine use, to a contractor of the Agency having need for the information in order to perform a contract. Recipients of information shall be required to comply with the requirements of the Privacy Act of 1974, as amended, pursuant to 5 U.S.C. 552a(m).
5. A record related to an International Application filed under the Patent Cooperation Treaty in this system of records may be disclosed, as a routine use, to the International Bureau of the World Intellectual Property Organization, pursuant to the Patent Cooperation Treaty.
6. A record in this system of records may be disclosed, as a routine use, to another federal agency for purposes of National Security review (35 U.S.C. 181) and for review pursuant to the Atomic Energy Act (42 U.S.C. 218(c)).
7. A record from this system of records may be disclosed, as a routine use, to the Administrator, General Services, or his/her designee, during an inspection of records conducted by GSA as part of that agency's responsibility to recommend improvements in records management practices and programs, under authority of 44 U.S.C. 2904 and 2906. Such disclosure shall be made in accordance with the GSA regulations governing inspection of records for this purpose, and any other relevant (i.e., GSA or Commerce) directive. Such disclosure shall not be used to make determinations about individuals.
8. A record from this system of records may be disclosed, as a routine use, to the public after either publication of the application pursuant to 35 U.S.C. 122(b) or issuance of a patent pursuant to 35 U.S.C. 151. Further, a record may be disclosed, subject to the limitations of 37 CFR 1.14, as a routine use, to the public if the record was filed in an application which became abandoned or in which the proceedings were terminated and which application is referenced by either a published application, an application open to public inspection or an issued patent.
9. A record from this system of records may be disclosed, as a routine use, to a Federal, State, or local law enforcement agency, if the USPTO becomes aware of a violation or potential violation of law or regulation.

Notice of Allowability	Application No. 16/262,902	Applicant(s) Burgo et al.	
	Examiner NANNETTE HOLLOMAN	Art Unit 1612	AIA (FITF) Status Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address--

All claims being allowable, PROSECUTION ON THE MERITS IS (OR REMAINS) CLOSED in this application. If not included herewith (or previously mailed), a Notice of Allowance (PTOL-85) or other appropriate communication will be mailed in due course. **THIS NOTICE OF ALLOWABILITY IS NOT A GRANT OF PATENT RIGHTS.** This application is subject to withdrawal from issue at the initiative of the Office or upon petition by the applicant. See 37 CFR 1.313 and MPEP 1308.

1. ☑ This communication is responsive to amendments filed 06/03/2022.
   ☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____.

2. ☐ An election was made by the applicant in response to a restriction requirement set forth during the interview on _____; the restriction requirement and election have been incorporated into this action.

3. ☑ The allowed claim(s) is/are 88-93 and 95-111 . As a result of the allowed claim(s), you may be eligible to benefit from the **Patent Prosecution Highway** program at a participating intellectual property office for the corresponding application. For more information , please see **http://www.uspto.gov/patents/init_events/pph/index.jsp** or send an inquiry to **PPHfeedback@uspto.gov.**

4. ☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

   Certified copies:

   a) ☐ All b) ☐ Some* c) ☐ None of the:

   1. ☐ Certified copies of the priority documents have been received.
   2. ☐ Certified copies of the priority documents have been received in Application No. _____ .
   3. ☐ Copies of the certified copies of the priority documents have been received in this national stage application from the International Bureau (PCT Rule 17.2(a)).

   * Certified copies not received: _____ .

Applicant has THREE MONTHS FROM THE "MAILING DATE" of this communication to file a reply complying with the requirements noted below. Failure to timely comply will result in ABANDONMENT of this application.
THIS THREE-MONTH PERIOD IS NOT EXTENDABLE.

5. ☐ CORRECTED DRAWINGS (as "replacement sheets") must be submitted.
   ☐ including changes required by the attached Examiner's Amendment / Comment or in the Office action of Paper No./Mail Date _____ .

   Identifying indicia such as the application number (see 37 CFR 1.84(c)) should be written on the drawings in the front (not the back) of each sheet. Replacement sheet(s) should be labeled as such in the header according to 37 CFR 1.121(d).

6. ☐ DEPOSIT OF and/or INFORMATION about the deposit of BIOLOGICAL MATERIAL must be submitted. Note the attached Examiner's comment regarding REQUIREMENT FOR THE DEPOSIT OF BIOLOGICAL MATERIAL.

Attachment(s)

1. ☐ Notice of References Cited (PTO-892)
2. ☐ Information Disclosure Statements (PTO/SB/08), Paper No./Mail Date _____.
3. ☐ Examiner's Comment Regarding Requirement for Deposit of Biological Material _____.
4. ☑ Interview Summary (PTO-413), Paper No./Mail Date. _____.
5. ☑ Examiner's Amendment/Comment
6. ☐ Examiner's Statement of Reasons for Allowance
7. ☐ Other _____.

/NANNETTE HOLLOMAN/
Primary Examiner, Art Unit 1612

6889441Notice of Pre-AIA or AIA Status

The present application, filed on or after March 16, 2013, is being examined under the first inventor to file provisions of the AIA.

EXAMINER'S AMENDMENT

An examiner's amendment to the record appears below. Should the changes and/or additions be unacceptable to applicant, an amendment may be filed as provided by 37 CFR 1.312. To ensure consideration of such an amendment, it MUST be submitted no later than the payment of the issue fee.

Authorization for this examiner's amendment was given in an interview with Aaron Lukas on 06/16/2022.

The application has been amended as follows:

Claim 95, line 1, the phrase "claim 94" is replaced with --- claim 93 ---.

Claim 96, line 1, the phrase "claim 94" is replaced with --- claim 93 ---.

Claim 96, line 4, the term "bio-1,2-hexanediol" is deleted.

Claim 101, line 4, the term "bio-1,2-hexanediol" is deleted.

Claim 104, line 4, the term "bio-1,2-hexanediol" is deleted.

Claim 94 is cancelled.

Conclusion

Any inquiry concerning this communication or earlier communications from the examiner should be directed to NANNETTE HOLLOMAN whose telephone number is (571)270-5231. The examiner can normally be reached Monday-Friday 9am-6pm.

Examiner interviews are available via telephone, in-person, and video conferencing using a USPTO supplied web-based collaboration tool. To schedule an interview, applicant is encouraged to use the USPTO Automated Interview Request (AIR) at http://www.uspto.gov/interviewpractice.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Frederick Krass can be reached on (571)272-0580. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of published or unpublished applications may be obtained from Patent Center. Unpublished application information in Patent Center is available to registered users. To file and manage patent submissions in Patent Center, visit: https://patentcenter.uspto.gov. Visit https://www.uspto.gov/patents/apply/patent-center for more information about Patent Center and https://www.uspto.gov/patents/docx for information about filing in DOCX format. For additional questions, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

/NANNETTE HOLLOMAN/
Primary Examiner, Art Unit 1612

Examiner-Initiated Interview Summary	**Application No.** 16/262,902	**Applicant(s)** Burgo et al.		
	Examiner NANNETTE HOLLOMAN	**Art Unit** 1612	**AIA (First Inventor to File) Status** Yes	**Page** 1 of 1

All Participants (applicant, applicants representative, PTO personnel)	**Title**	**Type**
NANNETTE HOLLOMAN	Primary Examiner	Telephonic
AARON LUKAS	Attorney of Record	

Date of Interview: 16 June 2022

Issues Discussed:

Proposed Amendment(s)

Proposed amendments to delete new matter and amend claim dependencies. An agreement was made and the claim amendments are presented in the attached examiner's amendment.

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Applicant is reminded that a complete written statement as to the substance of the interview must be made of record in the application file. It is the applicants responsibility to provide the written statement, unless the interview was initiated by the Examiner and the Examiner has indicated that a written summary will be provided. See MPEP 713.04
Please further see:
MPEP 713.04
Title 37 Code of Federal Regulations (CFR) § 1.133 Interviews, paragraph (b)
37 CFR § 1.2 Business to be transacted in writing

Applicant recordation instructions: It is not necessary for applicant to provide a separate record of the substance of interview.

Examiner recordation instructions: Examiners must summarize the substance of any interview of record. A complete and proper recordation of the substance of an interview should include the items listed in MPEP 713.04 for complete and proper recordation including the identification of the general thrust of each argument or issue discussed, a general indication of any other pertinent matters discussed regarding patentability and the general results or outcome of the interview, to include an indication as to whether or not agreement was reached on the issues raised.

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	**Art Unit**
	NANNETTE HOLLOMAN	1612

CPC								
Symbol							**Type**	**Version**
C07C	/	29	/	04			F	2013-01-01
C07C	/	31	/	20			I	2013-01-01
C07C	/	29	/	80			I	2013-01-01
B01J	/	23	/	06			I	2013-01-01
B01J	/	37	/	08			I	2013-01-01
B01J	/	21	/	04			I	2013-01-01
C11D	/	3	/	2044			I	2013-01-01
C07C	/	1	/	24			I	2013-01-01
C07C	/	29	/	48			I	2013-01-01
C07C	/	11	/	02			I	2013-01-01
C07C	/	11	/	107			I	2013-01-01
B01J	/	2523	/	27			A	2013-01-01
C07B	/	41	/	02			A	2013-01-01
C07B	/	63	/	00			A	2013-01-01
B01J	/	2523	/	31			A	2013-01-01
A61L	/	2	/	18			A	2013-01-01
A01N	/	31	/	02			A	2013-01-01
A61K	/	8	/	345			A	2013-01-01
A61Q	/	5	/	00			A	2013-01-01
A61Q	/	11	/	00			A	2013-01-01
A61Q	/	19	/	00			A	2013-01-01
C11D	/	3	/	48			A	2013-01-01
C07C	/	2521	/	04			A	2013-01-01

CPC Combination Sets									
Symbol							**Type**	**Set**	**Ranking**	**Version**
C07C	/	1	/	24			I	1	1	2013-01-01
C07C	/	11	/	02			I	1	2	2013-01-01
C07C	/	29	/	48			I	2	1	2013-01-01
C07C	/	31	/	20			I	2	2	2013-01-01

NONE		**Total Claims Allowed:**	
(Assistant Examiner)	(Date)	23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	16 June 2022	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	NONE

U.S. Patent and Trademark Office — Part of Paper No.: 20220616

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	**Art Unit**
	NANNETTE HOLLOMAN	1612

CPC Combination Sets								
Symbol					**Type**	**Set**	**Ranking**	**Version**
C07C	/	29	/	80	I	3	1	2013-01-01
C07C	/	31	/	20	I	3	2	2013-01-01

NONE		**Total Claims Allowed:**	
(Assistant Examiner)	(Date)	23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	16 June 2022	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	NONE

U.S. Patent and Trademark Office

Part of Paper No.: 20220616

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	**Art Unit**
	NANNETTE HOLLOMAN	1612

INTERNATIONAL CLASSIFICATION					
CLAIMED					
C07C29/04	/	29	/	04	

NON-CLAIMED					
	/		/		

US ORIGINAL CLASSIFICATION	
CLASS	**SUBCLASS**

CROSS REFERENCES(S)						
CLASS	**SUBCLASS (ONE SUBCLASS PER BLOCK)**					

NONE (Assistant Examiner) (Date)	**Total Claims Allowed:** 23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612 (Primary Examiner) 16 June 2022 (Date)	O.G. Print Claim(s) 1	O.G. Print Figure NONE

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	**Art Unit**
	NANNETTE HOLLOMAN	1612

☐ Claims renumbered in the same order as presented by applicant ☐ CPA ☐ T.D. ☐ R.1.47

CLAIMS

Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original
	1		15		29		43		57		71		85	11	99
	2		16		30		44		58		72		86	12	100
	3		17		31		45		59		73		87	13	101
	4		18		32		46		60		74	1	88	14	102
	5		19		33		47		61		75	2	89	15	103
	6		20		34		48		62		76	3	90	16	104
	7		21		35		49		63		77	4	91	17	105
	8		22		36		50		64		78	5	92	18	106
	9		23		37		51		65		79	6	93	19	107
	10		24		38		52		66		80		94	20	108
	11		25		39		53		67		81	7	95	21	109
	12		26		40		54		68		82	8	96	22	110
	13		27		41		55		69		83	9	97	23	111
	14		28		42		56		70		84	10	98		

NONE (Assistant Examiner)	(Date)	**Total Claims Allowed:** 23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612 (Primary Examiner)	16 June 2022 (Date)	O.G. Print Claim(s) 1	O.G. Print Figure NONE

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

CPC - Searched*		
Symbol	**Date**	**Examiner**
A01N	04/10/2021	NH
C07C	04/10/2021	NH
A61K	04/10/2021	NH
A61Q	04/10/2021	NH

CPC Combination Sets - Searched*		
Symbol	**Date**	**Examiner**

US Classification - Searched*			
Class	**Subclass**	**Date**	**Examiner**

* See search history printout included with this form or the SEARCH NOTES box below to determine the scope of the search.

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

Search Notes		
Search Notes	**Date**	**Examiner**
PALM Inventor Search	09/13/2020	NH
EAST Search	09/13/2020	NH
NPL Search	09/13/2020	NH
PALM Inventor Search	08/26/2021	NH
EAST Search	08/26/2021	NH
NPL Search	08/26/2021	NH
PALM Inventor Search	03/25/2022	NH
EAST Search	03/25/2022	NH
NPL Search	03/25/2022	NH
PALM Inventor Search	06/16/2022	NH
SEARCH	06/16/2022	NH
NPL Search	06/16/2022	NH

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

Interference Search			
US Class/CPC Symbol	**US Subclass/CPC Group**	**Date**	**Examiner**
C07C	11/18, 2527/054, 2527/167, 2527/173, 2531/02, 65/21,	04/10/2021	NH
A61K	8/345, 2300/00, 2800/31, 8/60, 2800/412, 8/11, 8/732, 8/738, 31/197, 8/44, 8/73, 2800/48, 8/25, 8/34, 8/585, 8/92, 8/9789, 9/014, 2800/30, 31/198, 36/28, 36/534, 36/67	04/10/2021	NH
A61Q	15/00, 19/00, 13/00, 11/00, 5/00, 19/007, 19/08, 19/02, 17/005, 17/04, 19/10, 5/02, 7/00	04/10/2021	NH
C07C	11/18, 2527/054, 2527/167, 2527/173, 2531/02, 65/21,	06/16/2022	NH
A61K	8/345, 2300/00, 2800/31, 8/60, 2800/412, 8/11, 8/732, 8/738, 31/197, 8/44, 8/73, 2800/48, 8/25, 8/34, 8/585, 8/92, 8/9789, 9/014, 2800/30, 31/198, 36/28, 36/534, 36/67	06/16/2022	NH
A61Q	15/00, 19/00, 13/00, 11/00, 5/00, 19/007, 19/08, 19/02, 17/005, 17/04, 19/10, 5/02, 7/00	06/16/2022	NH

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

PE2E SEARCH - Search History (Prior Art)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	British Equivalents	Time Stamp
L1	937	((("BURGO") near3 ("Rocco")) OR (("WRIGHT") near3 ("Michael")) OR (("MOSSER") near3 ("Gary")) OR (("FEVOLA") near3 ("Michael"))).INV.	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	OFF	OFF	2020/07/07 07:12 PM
L2	378	(bio with alkene) bioalkene	(US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT)	OR	ON	ON	2020/07/07 07:19 PM
L3	6577	regioselectivity	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	ON	ON	2020/09/09 02:30 PM
L4	13	regioselectivity same alkanediol	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	ON	ON	2020/09/09 02:30 PM
L5	7	regioselectivity same (octanediol or (caprylyl adj glycol))	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	ON	ON	2020/09/09 02:47 PM
L6	1	("7385092").PN.	(USPAT; USOCR)	OR	OFF	OFF	2020/09/09 02:53 PM
L7	2	wo-2016007196-$.did.	(US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT)	OR	OFF	OFF	2020/09/11 11:19 AM
L8	6	alkanediol same (household and pharmaceutical and cosmetic)	(US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT)	OR	ON	ON	2020/09/11 05:25 PM
L9	3	(household and pharmaceutical and cosmetic) same (octanediol or (caprylyl adj glycol))	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	ON	ON	2020/09/11 05:26 PM
L10	336	(household and pharmaceutical and cosmetic) and (octanediol or (caprylyl adj glycol))	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	ON	ON	2020/09/11 05:26 PM
L11	6	((household and pharmaceutical and cosmetic) and (octanediol or (caprylyl adj glycol))).clm.	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	ON	ON	2020/09/11 05:26 PM
L12	11	((household and pharmaceutical and cosmetic) and (octanediol or ((caprylyl or hexylene or	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	ON	ON	2020/09/11 05:47 PM

		decylene) adj glycol) or alkanediol)).clm.					
L13	946	((("BURGO") near3 ("Rocco")) OR (("WRIGHT") near3 ("Michael")) OR (("MOSSER") near3 ("Gary")) OR (("FEVOLA") near3 ("Michael"))).INV.	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	OFF	OFF	2020/09/11 07:14 PM
L14	3	L13 and alkanediol	(US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT)	OR	ON	ON	2020/09/11 07:18 PM
L15	7	(("8912373") or ("9856461") or ("4234752") or ("20050065389") or ("20030065233") or ("20060161025") or ("20140066666")).PN.	(US-PGPUB; USPAT; USOCR)	OR	OFF	OFF	2020/09/13 01:37 PM
L16	42261	decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	ON	ON	2021/02/27 11:31 PM
L17	13209	L16 and (preservative or pharmaceutical or cosmetic)	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	ON	ON	2021/02/27 11:32 PM
L18	1488	L17 and "bio"	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	ON	ON	2021/02/27 11:32 PM
L19	67	"bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol)))	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	ON	ON	2021/04/10 07:56 PM
L20	549665	((A01N47/44 OR A01N37/36 OR A01N59/16 OR A01N31/02 OR A01N25/34 OR A01N59/00 OR A01N61/00 OR A01N25/00 OR A01N2300/00 OR A01N25/10 OR C07C31/20 OR C07C1/24 OR C07C11/02 OR C07C29/48 OR C07C29/80 OR C07C263/12 OR C07C265/14 OR C07C2521/04 OR C07C11/107 OR C07C29/04 OR C07C247/24 OR	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	ON	ON	2021/04/10 07:57 PM

		C07C11/167 OR C07C11/18 OR C07C2527/054 OR C07C2527/167 OR C07C2527/173 OR C07C2531/02 OR C07C65/21 OR A61K8/345 OR A61K2300/00 OR A61K2800/31 OR A61K8/60 OR A61K2800/412 OR A61K8/11 OR A61K8/732 OR A61K8/738 OR A61K31/197 OR A61K8/44 OR A61K8/73 OR A61K2800/48 OR A61K8/25 OR A61K8/34 OR A61K8/585 OR A61K8/92 OR A61K8/9789 OR A61K9/0014 OR A61K2800/30 OR A61K31/198 OR A61K36/28 OR A61K36/534 OR A61K36/67 OR A61K36/886 OR A61K45/06 OR A61K47/10 OR A61K8/24 OR A61K8/362 OR A61K8/37 OR A61K8/41 OR A61K8/42 OR A61K8/4973 OR A61K8/498 OR A61K8/891 OR A61K9/06 OR A61K2800/10 OR A61K2800/28 OR A61K2800/524 OR A61K2800/56 OR A61K2800/87 OR A61K2800/874 OR A61K31/327 OR A61K47/22 OR A61K47/44 OR A61K47/46 OR A61K8/0216 OR A61K8/046 OR A61K8/19 OR A61K8/20 OR A61K8/23 OR A61K8/26 OR					

		A61K8/31 OR A61K8/64 OR A61K8/678 OR A61K8/735 OR A61K8/8111 OR A61K8/8147 OR A61K8/893 OR A61K8/895 OR A61K8/922 OR A61K8/9794 OR A61K9/107 OR A61K2035/115 OR A61K2800/43 OR A61K2800/522 OR A61K2800/592 OR A61K2800/5922 OR A61K2800/594 OR A61K2800/596 OR A61K2800/624 OR A61K2800/805 OR A61K31/192 OR A61K31/355 OR A61K31/60 OR A61K33/30 OR A61K35/745 OR A61K35/747 OR A61K36/185 OR A61K36/53 OR A61K47/02 OR A61K47/06 OR A61K47/12 OR A61K47/14 OR A61K47/183 OR A61K47/24 OR A61K47/34 OR A61K47/38 OR A61K49/0006 OR A61K49/0008 OR A61K8/06 OR A61K8/062 OR A61K8/28 OR A61K8/29 OR A61K8/342 OR A61K8/347 OR A61K8/365 OR A61K8/368 OR A61K8/375 OR A61K8/416 OR A61K8/445 OR A61K8/463 OR A61K8/49 OR A61K8/4926 OR A61K8/63 OR A61K8/66 OR A61K8/675 OR A61K8/8152 OR A61K8/8194 OR A61K8/927 OR					

		A61K8/965 OR A61K8/97 OR A61K9/08 OR A61Q15/00 OR A61Q19/00 OR A61Q13/00 OR A61Q11/00 OR A61Q5/00 OR A61Q19/007 OR A61Q19/08 OR A61Q19/02 OR A61Q17/005 OR A61Q17/04 OR A61Q19/10 OR A61Q5/02 OR A61Q7/00).CPC.)					
L21	42	L20 and ("bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))))	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	ON	ON	2021/04/10 07:57 PM
L24	2	(("9242226") or ("20050222276")).PN.	(US-PGPUB; USPAT; USOCR)	OR	OFF	OFF	2021/08/14 09:51 PM
L25	4	("20050069997"|"7385092"|"8450543"|"9732295").PN.	(US-PGPUB; USPAT)	OR	OFF	OFF	2021/11/26 11:34 AM
L26	4	("20050069997"|"7385092"|"8450543"|"9732295").PN.	(US-PGPUB; USPAT)	OR	OFF	OFF	2021/11/26 11:35 AM
L27	48937	"bio".ti.	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	ON	ON	2022/01/19 12:50 PM
L28	43	(natural or (natural adj derived)) with "1,2-alkanediol"	(US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT)	OR	ON	ON	2022/01/19 12:51 PM
L29	2	(("8912373") or ("9242226")).PN.	(USPAT; USOCR)	OR	OFF	OFF	2022/01/19 12:59 PM
L30	3	("20090004122"|"20090143489"|"20120238788").PN.	(US-PGPUB; USPAT)	OR	OFF	OFF	2022/03/23 08:39 PM
L31	993	((("BURGO") near3 ("Rocco")) OR (("WRIGHT") near3 ("Michael")) OR (("MOSSER") near3 ("Gary")) OR (("FEVOLA") near3 ("Michael"))).INV.	(US-PGPUB; USPAT; USOCR; EPO; JPO; DERWENT)	OR	OFF	OFF	2022/03/25 02:21 PM
L32	6	L31 and alkanediol	(US-PGPUB; USPAT; FPRS; EPO; JPO; DERWENT)	OR	ON	ON	2022/03/25 02:21 PM
L33	7	L31 AND alkanediol	(US-PGPUB; USPAT)	AND	ON	ON	2022/06/16 01:40 PM

PE2E SEARCH - Search History (Interference)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	British Equivalents	Time Stamp
N22	125400	((A01N47/44 OR A01N37/36 OR A01N59/16 OR A01N31/02 OR A01N25/34 OR A01N59/00 OR A01N61/00 OR A01N25/00 OR A01N2300/00 OR A01N25/10 OR C07C31/20 OR C07C1/24 OR C07C11/02 OR C07C29/48 OR C07C29/80 OR C07C263/12 OR C07C265/14 OR C07C2521/04 OR C07C11/107 OR C07C29/04 OR C07C247/24 OR C07C11/167 OR C07C11/18 OR C07C2527/054 OR C07C2527/167 OR C07C2527/173 OR C07C2531/02 OR C07C65/21 OR A61K8/345 OR A61K2300/00 OR A61K2800/31 OR A61K8/60 OR A61K2800/412 OR A61K8/11 OR A61K8/732 OR A61K8/738 OR A61K31/197 OR A61K8/44 OR A61K8/73 OR A61K2800/48 OR A61K8/25 OR A61K8/34 OR A61K8/585 OR A61K8/92 OR A61K8/9789 OR A61K9/0014 OR A61K2800/30 OR A61K31/198 OR A61K36/28 OR A61K36/534 OR A61K36/67 OR A61K36/886 OR A61K45/06 OR	(USPAT)	OR	ON	ON	2021/04/10 07:59 PM

		A61K47/10 OR A61K8/24 OR A61K8/362 OR A61K8/37 OR A61K8/41 OR A61K8/42 OR A61K8/4973 OR A61K8/498 OR A61K8/891 OR A61K9/06 OR A61K2800/10 OR A61K2800/28 OR A61K2800/524 OR A61K2800/56 OR A61K2800/87 OR A61K2800/874 OR A61K31/327 OR A61K47/22 OR A61K47/44 OR A61K47/46 OR A61K8/0216 OR A61K8/046 OR A61K8/19 OR A61K8/20 OR A61K8/23 OR A61K8/26 OR A61K8/31 OR A61K8/64 OR A61K8/678 OR A61K8/735 OR A61K8/8111 OR A61K8/8147 OR A61K8/893 OR A61K8/895 OR A61K8/922 OR A61K8/9794 OR A61K9/107 OR A61K2035/115 OR A61K2800/43 OR A61K2800/522 OR A61K2800/592 OR A61K2800/5922 OR A61K2800/594 OR A61K2800/596 OR A61K2800/624 OR A61K2800/805 OR A61K31/192 OR A61K31/355 OR A61K31/60 OR A61K33/30 OR A61K35/745 OR A61K35/747 OR A61K36/185 OR A61K36/53 OR A61K47/02 OR A61K47/06 OR A61K47/12 OR A61K47/14 OR					

		A61K47/183 OR A61K47/24 OR A61K47/34 OR A61K47/38 OR A61K49/0006 OR A61K49/0008 OR A61K8/06 OR A61K8/062 OR A61K8/28 OR A61K8/29 OR A61K8/342 OR A61K8/347 OR A61K8/365 OR A61K8/368 OR A61K8/375 OR A61K8/416 OR A61K8/445 OR A61K8/463 OR A61K8/49 OR A61K8/4926 OR A61K8/63 OR A61K8/66 OR A61K8/675 OR A61K8/8152 OR A61K8/8194 OR A61K8/927 OR A61K8/965 OR A61K8/97 OR A61K9/08 OR A61Q15/00 OR A61Q19/00 OR A61Q13/00 OR A61Q11/00 OR A61Q5/00 OR A61Q19/007 OR A61Q19/08 OR A61Q19/02 OR A61Q17/005 OR A61Q17/04 OR A61Q19/10 OR A61Q5/02 OR A61Q7/00).CPC.)					
N23	10	N22 and ("bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))))	(USPAT)	OR	ON	ON	2021/04/10 07:59 PM
N24	134090	((A01N47/44 OR A01N37/36 OR A01N59/16 OR A01N31/02 OR A01N25/34 OR A01N59/00 OR A01N61/00 OR A01N25/00 OR A01N2300/00 OR A01N25/10 OR	(USPAT)	OR	ON	ON	2022/06/16 01:41 PM

		C07C31/20 OR C07C1/24 OR C07C11/02 OR C07C29/48 OR C07C29/80 OR C07C263/12 OR C07C265/14 OR C07C2521/04 OR C07C11/107 OR C07C29/04 OR C07C247/24 OR C07C11/167 OR C07C11/18 OR C07C2527/054 OR C07C2527/167 OR C07C2527/173 OR C07C2531/02 OR C07C65/21 OR A61K8/345 OR A61K2300/00 OR A61K2800/31 OR A61K8/60 OR A61K2800/412 OR A61K8/11 OR A61K8/732 OR A61K8/738 OR A61K31/197 OR A61K8/44 OR A61K8/73 OR A61K2800/48 OR A61K8/25 OR A61K8/34 OR A61K8/585 OR A61K8/92 OR A61K8/9789 OR A61K9/0014 OR A61K2800/30 OR A61K31/198 OR A61K36/28 OR A61K36/534 OR A61K36/67 OR A61K36/886 OR A61K45/06 OR A61K47/10 OR A61K8/24 OR A61K8/362 OR A61K8/37 OR A61K8/41 OR A61K8/42 OR A61K8/4973 OR A61K8/498 OR A61K8/891 OR A61K9/06 OR A61K2800/10 OR A61K2800/28 OR A61K2800/524 OR A61K2800/56 OR A61K2800/87 OR					

		A61K2800/874 OR A61K31/327 OR A61K47/22 OR A61K47/44 OR A61K47/46 OR A61K8/0216 OR A61K8/046 OR A61K8/19 OR A61K8/20 OR A61K8/23 OR A61K8/26 OR A61K8/31 OR A61K8/64 OR A61K8/678 OR A61K8/735 OR A61K8/8111 OR A61K8/8147 OR A61K8/893 OR A61K8/895 OR A61K8/922 OR A61K8/9794 OR A61K9/107 OR A61K2035/115 OR A61K2800/43 OR A61K2800/522 OR A61K2800/592 OR A61K2800/5922 OR A61K2800/594 OR A61K2800/596 OR A61K2800/624 OR A61K2800/805 OR A61K31/192 OR A61K31/355 OR A61K31/60 OR A61K33/30 OR A61K35/745 OR A61K35/747 OR A61K36/185 OR A61K36/53 OR A61K47/02 OR A61K47/06 OR A61K47/12 OR A61K47/14 OR A61K47/183 OR A61K47/24 OR A61K47/34 OR A61K47/38 OR A61K49/0006 OR A61K49/0008 OR A61K8/06 OR A61K8/062 OR A61K8/28 OR A61K8/29 OR A61K8/342 OR A61K8/347 OR A61K8/365 OR A61K8/368 OR A61K8/375 OR				

		A61K8/416 OR A61K8/445 OR A61K8/463 OR A61K8/49 OR A61K8/4926 OR A61K8/63 OR A61K8/66 OR A61K8/675 OR A61K8/8152 OR A61K8/8194 OR A61K8/927 OR A61K8/965 OR A61K8/97 OR A61K9/08 OR A61Q15/00 OR A61Q19/00 OR A61Q13/00 OR A61Q11/00 OR A61Q5/00 OR A61Q19/007 OR A61Q19/08 OR A61Q19/02 OR A61Q17/005 OR A61Q17/04 OR A61Q19/10 OR A61Q5/02 OR A61Q7/00).CPC.)					
N25	22	N22 and ("bio" with (decanediol or dodecanediol or (octanediol or (caprylyl adj glycol))))	(USPAT)	OR	ON	ON	2022/06/16 01:41 PM

PART B - FEE(S) TRANSMITTAL

Complete and send this form, together with applicable fee(s), by mail or fax, or via EFS-Web.

By mail, send to: Mail Stop ISSUE FEE
Commissioner for Patents
P.O. Box 1450
Alexandria, Virginia 22313-1450

By fax, send to: (571)-273-2885

INSTRUCTIONS: This form should be used for transmitting the ISSUE FEE and PUBLICATION FEE (if required). Blocks 1 through 5 should be completed where appropriate. All further correspondence including the Patent, advance orders and notification of maintenance fees will be mailed to the current correspondence address as indicated unless corrected below or directed otherwise in Block 1, by (a) specifying a new correspondence address; and/or (b) indicating a separate "FEE ADDRESS" for maintenance fee notifications.

CURRENT CORRESPONDENCE ADDRESS (Note: Use Block 1 for any change of address)

171052 7590 06/23/2022

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

Note: A certificate of mailing can only be used for domestic mailings of the Fee(s) Transmittal. This certificate cannot be used for any other accompanying papers. Each additional paper, such as an assignment or formal drawing, must have its own certificate of mailing or transmission.

Certificate of Mailing or Transmission

I hereby certify that this Fee(s) Transmittal is being deposited with the United States Postal Service with sufficient postage for first class mail in an envelope addressed to the Mail Stop ISSUE FEE address above, or being transmitted to the USPTO via EFS-Web or by facsimile to (571) 273-2885, on the date below.

(Typed or printed name)
(Signature)
(Date)

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	INLX.0198U2/00484799	1028

TITLE OF INVENTION: Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same

APPLN. TYPE	ENTITY STATUS	ISSUE FEE DUE	PUBLICATION FEE DUE	PREV. PAID ISSUE FEE	TOTAL FEE(S) DUE	DATE DUE
nonprovisional	UNDISCOUNTED	$1200	$0.00	$0.00	$1200	09/23/2022

EXAMINER	ART UNIT	CLASS-SUBCLASS
HOLLOMAN, NANNETTE	1612	424-049000

1. Change of correspondence address or indication of "Fee Address" (37 CFR 1.363).

☐ Change of correspondence address (or Change of Correspondence Address form PTO/AIA/122 or PTO/SB/122) attached.

☐ "Fee Address" indication (or "Fee Address" Indication form PTO/AIA/47 or PTO/SB/47; Rev 03-02 or more recent) attached. **Use of a Customer Number is required.**

2. For printing on the patent front page, list
(1) The names of up to 3 registered patent attorneys or agents OR, alternatively,
(2) The name of a single firm (having as a member a registered attorney or agent) and the names of up to 2 registered patent attorneys or agents. If no name is listed, no name will be printed.

1 _____
2 COZEN O'CONNOR
3 _____

3. ASSIGNEE NAME AND RESIDENCE DATA TO BE PRINTED ON THE PATENT (print or type)

PLEASE NOTE: Unless an assignee is identified below, no assignee data will appear on the patent. If an assignee is identified below, the document must have been previously recorded, or filed for recordation, as set forth in 37 CFR 3.11 and 37 CFR 3.81(a). Completion of this form is NOT a substitute for filing an assignment.

(A) NAME OF ASSIGNEE: INOLEX INVESTMENT CORPORATION

(B) RESIDENCE: (CITY and STATE OR COUNTRY): WILMINGTON, DELAWARE

Please check the appropriate assignee category or categories (will not be printed on the patent) : ☐ Individual ☒ Corporation or other private group entity ☐ Government

4a. Fees submitted: ☒ Issue Fee ☐ Publication Fee (if required) ☐ Advance Order - # of Copies _____

4b. Method of Payment: *(Please first reapply any previously paid fee shown above)*

☒ Electronic Payment via EFS-Web ☐ Enclosed check ☐ Non-electronic payment by credit card (Attach form PTO-2038)

☒ The Director is hereby authorized to charge the required fee(s), any deficiency, or credit any overpayment to Deposit Account No. 50-3111

5. **Change in Entity Status** (from status indicated above)

☐ Applicant certifying micro entity status. See 37 CFR 1.29

☐ Applicant asserting small entity status. See 37 CFR 1.27

☐ Applicant changing to regular undiscounted fee status.

NOTE: Absent a valid certification of Micro Entity Status (see forms PTO/SB/15A and 15B), issue fee payment in the micro entity amount will not be accepted at the risk of application abandonment.
NOTE: If the application was previously under micro entity status, checking this box will be taken to be a notification of loss of entitlement to micro entity status.
NOTE: Checking this box will be taken to be a notification of loss of entitlement to small or micro entity status, as applicable.

NOTE: This form must be signed in accordance with 37 CFR 1.31 and 1.33. See 37 CFR 1.4 for signature requirements and certifications.

Authorized Signature */Aaron S. Lukas, Reg. #59,443/* Date June 24, 2022

Typed or printed name Aaron S. Lukas, Ph.D. Registration No. 59,443

Electronic Patent Application Fee Transmittal

Application Number:	16262902
Filing Date:	30-Jan-2019
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Filer:	Aaron Scott Lukas/Valerie Foster
Attorney Docket Number:	INLX.0198U2/00484799

Filed as Large Entity

Filing Fees for Utility under 35 USC 111(a)

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Pages:				
Claims:				
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
UTILITY APPL ISSUE FEE	1501	1	1200	1200

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Extension-of-Time:				
Miscellaneous:				
			Total in USD ($)	**1200**

Electronic Acknowledgement Receipt

EFS ID:	46037586
Application Number:	16262902
International Application Number:	
Confirmation Number:	1028
Title of Invention:	Natural 1,2-Alkanediols, Compositions Having Natural 1,2-Alkanediols and Processes for Making the Same
First Named Inventor/Applicant Name:	Rocco V. Burgo
Customer Number:	171052
Filer:	Aaron Scott Lukas/Valerie Foster
Filer Authorized By:	Aaron Scott Lukas
Attorney Docket Number:	INLX.0198U2/00484799
Receipt Date:	24-JUN-2022
Filing Date:	30-JAN-2019
Time Stamp:	12:14:50
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	CARD
Payment was successfully received in RAM	$1200
RAM confirmation Number	E20226NC15174262
Deposit Account	
Authorized User	

The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows:

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Issue Fee Payment (PTO-85B)	INLX_0198U2_00484799_IssueFeeTransmittal.pdf	126063 d2f8faebf27ecf685d8f066e5cb3980b15ea3f55	no	1
Warnings:					
Information:					
2	Fee Worksheet (SB06)	fee-info.pdf	38279 bd9bb4c6107b3c59a5955a9c839578dda85949f4	no	2
Warnings:					
Information:					
		Total Files Size (in bytes):	164342		

This Acknowledgement Receipt evidences receipt on the noted date by the USPTO of the indicated documents, characterized by the applicant, and including page counts, where applicable. It serves as evidence of receipt similar to a Post Card, as described in MPEP 503.

New Applications Under 35 U.S.C. 111

If a new application is being filed and the application includes the necessary components for a filing date (see 37 CFR 1.53(b)-(d) and MPEP 506), a Filing Receipt (37 CFR 1.54) will be issued in due course and the date shown on this Acknowledgement Receipt will establish the filing date of the application.

National Stage of an International Application under 35 U.S.C. 371

If a timely submission to enter the national stage of an international application is compliant with the conditions of 35 U.S.C. 371 and other applicable requirements a Form PCT/DO/EO/903 indicating acceptance of the application as a national stage submission under 35 U.S.C. 371 will be issued in addition to the Filing Receipt, in due course.

New International Application Filed with the USPTO as a Receiving Office

If a new international application is being filed and the international application includes the necessary components for an international filing date (see PCT Article 11 and MPEP 1810), a Notification of the International Application Number and of the International Filing Date (Form PCT/RO/105) will be issued in due course, subject to prescriptions concerning national security, and the date shown on this Acknowledgement Receipt will establish the international filing date of the application.

Doc Code:IDS.3P
Document Description: Third-Party Submission Under 37 CFR 1.290

PTO/SB/429(08-12)
Approved for use through 07/31/2015. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it contains a valid OMB control number.

THIRD-PARTY SUBMISSION UNDER 37 CFR 1.290	Application Number	16262902

U.S. PATENTS

Cite No	Patent Number	Kind Code[1]	Issue Date (YYYY-MM-DD)	First Named Inventor
1	4234752		1980-11-18	YULIN WU

U.S. PATENT APPLICATION PUBLICATIONS

Cite No	Publication Number	Kind Code[1]	Publication Date (YYYY-MM-DD)	First Named Inventor
1	20050065389	A1	2005-03-24	De Bruyn
2	20030065233	A1	2003-04-03	Fuji
3	20060161025	A1	~~2008-06-10~~ 07/2006	~~Swem~~ Osterholt ; et al.

Change(s) applied to document,
/A.J./
4/30/2021

FOREIGN PATENTS AND PUBLISHED FOREIGN PATENT APPLICATIONS

Cite No	Foreign Document Number[3]	Country Code[2]	Kind Code[1]	Publication Date (YYYY-MM-DD)	Applicant, Patentee or First Named Inventor	T[5]
1	1876162	EP	A1	2008-01-09	Furuya	☐

NON-PATENT PUBLICATIONS (e.g., journal article, Office action)

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /N.H/

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	01/30/2019	Rocco V. Burgo	INLX.0198U2/00484799	1028

171052 7590 07/14/2022

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

EXAMINER
HOLLOMAN, NANNETTE

ART UNIT	PAPER NUMBER
1612	

NOTIFICATION DATE	DELIVERY MODE
07/14/2022	ELECTRONIC

Please find below and/or attached an Office communication concerning this application or proceeding.

The time period for reply, if any, is set in the attached communication.

Notice of the Office communication was sent electronically on above-indicated "Notification Date" to the following e-mail address(es):

nyipdocketing@cozen.com
vdotson@cozen.com

Notice of Allowability	Application No. 16/262,902	Applicant(s) Burgo et al.	
	Examiner NANNETTE HOLLOMAN	Art Unit 1612	AIA (FITF) Status Yes

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address--

All claims being allowable, PROSECUTION ON THE MERITS IS (OR REMAINS) CLOSED in this application. If not included herewith (or previously mailed), a Notice of Allowance (PTOL-85) or other appropriate communication will be mailed in due course. **THIS NOTICE OF ALLOWABILITY IS NOT A GRANT OF PATENT RIGHTS.** This application is subject to withdrawal from issue at the initiative of the Office or upon petition by the applicant. See 37 CFR 1.313 and MPEP 1308.

1. ☑ This communication is responsive to internal communication 07/01/2022.
   ☐ A declaration(s)/affidavit(s) under **37 CFR 1.130(b)** was/were filed on _____.

2. ☐ An election was made by the applicant in response to a restriction requirement set forth during the interview on _____; the restriction requirement and election have been incorporated into this action.

3. ☑ The allowed claim(s) is/are 88-93 and 95-111 . As a result of the allowed claim(s), you may be eligible to benefit from the **Patent Prosecution Highway** program at a participating intellectual property office for the corresponding application. For more information , please see **http://www.uspto.gov/patents/init_events/pph/index.jsp** or send an inquiry to **PPHfeedback@uspto.gov.**

4. ☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

   Certified copies:

   a) ☐ All b) ☐ Some* c) ☐ None of the:

   1. ☐ Certified copies of the priority documents have been received.
   2. ☐ Certified copies of the priority documents have been received in Application No. _____ .
   3. ☐ Copies of the certified copies of the priority documents have been received in this national stage application from the International Bureau (PCT Rule 17.2(a)).

   * Certified copies not received: _____ .

Applicant has THREE MONTHS FROM THE "MAILING DATE" of this communication to file a reply complying with the requirements noted below. Failure to timely comply will result in ABANDONMENT of this application.
THIS THREE-MONTH PERIOD IS NOT EXTENDABLE.

5. ☐ CORRECTED DRAWINGS (as "replacement sheets") must be submitted.
   ☐ including changes required by the attached Examiner's Amendment / Comment or in the Office action of Paper No./Mail Date _____ .

   Identifying indicia such as the application number (see 37 CFR 1.84(c)) should be written on the drawings in the front (not the back) of each sheet. Replacement sheet(s) should be labeled as such in the header according to 37 CFR 1.121(d).

6. ☐ DEPOSIT OF and/or INFORMATION about the deposit of BIOLOGICAL MATERIAL must be submitted. Note the attached Examiner's comment regarding REQUIREMENT FOR THE DEPOSIT OF BIOLOGICAL MATERIAL.

Attachment(s)

1. ☐ Notice of References Cited (PTO-892)
2. ☐ Information Disclosure Statements (PTO/SB/08), Paper No./Mail Date _____.
3. ☐ Examiner's Comment Regarding Requirement for Deposit of Biological Material _____.
4. ☐ Interview Summary (PTO-413), Paper No./Mail Date. _____.
5. ☑ Examiner's Amendment/Comment
6. ☐ Examiner's Statement of Reasons for Allowance
7. ☐ Other _____.

/NANNETTE HOLLOMAN/
Primary Examiner, Art Unit 1612

Notice of Pre-AIA or AIA Status

The present application, filed on or after March 16, 2013, is being examined under the first inventor to file provisions of the AIA.

EXAMINER'S AMENDMENT

An examiner's amendment to the record appears below. Should the changes and/or additions be unacceptable to applicant, an amendment may be filed as provided by 37 CFR 1.312. To ensure consideration of such an amendment, it MUST be submitted no later than the payment of the issue fee.

Authorization for this examiner's amendment was given in an interview with Aaron Lukas on 06/16/2022.

The application has been amended as follows:

Claim 95, line 1, the phrase "claim 94" is replaced with --- claim 93 ---.

Claim 96, line 1, the phrase "claim 94" is replaced with --- claim 93 ---.

Claim 96, line 4, the term "bio-1,2-hexanediol" is deleted.

Claim 101, line 4, the term "bio-1,2-hexanediol" is deleted.

Claim 104, line 4, the term "bio-1,2-hexanediol" is deleted.

Claim 107, line 1, the phrase "claim 94" is replaced with --- claim 93 ---.

Claim 94 is cancelled.

Conclusion

Any inquiry concerning this communication or earlier communications from the examiner should be directed to NANNETTE HOLLOMAN whose telephone number is (571)270-5231. The examiner can normally be reached Monday-Friday 9am-6pm.

Examiner interviews are available via telephone, in-person, and video conferencing using a USPTO supplied web-based collaboration tool. To schedule an interview, applicant is encouraged to use the USPTO Automated Interview Request (AIR) at http://www.uspto.gov/interviewpractice.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Frederick Krass can be reached on (571)272-0580. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of published or unpublished applications may be obtained from Patent Center. Unpublished application information in Patent Center is available to registered users. To file and manage patent submissions in Patent Center, visit: https://patentcenter.uspto.gov. Visit https://www.uspto.gov/patents/apply/patent-center for more information about Patent Center and https://www.uspto.gov/patents/docx for information about filing in DOCX format. For additional questions, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

/NANNETTE HOLLOMAN/
Primary Examiner, Art Unit 1612

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

CPC - Searched*		
Symbol	**Date**	**Examiner**
A01N	04/10/2021	NH
C07C	04/10/2021	NH
A61K	04/10/2021	NH
A61Q	04/10/2021	NH

CPC Combination Sets - Searched*		
Symbol	**Date**	**Examiner**

US Classification - Searched*			
Class	**Subclass**	**Date**	**Examiner**

* See search history printout included with this form or the SEARCH NOTES box below to determine the scope of the search.

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

Search Notes		
Search Notes	**Date**	**Examiner**
PALM Inventor Search	09/13/2020	NH
EAST Search	09/13/2020	NH
NPL Search	09/13/2020	NH
PALM Inventor Search	08/26/2021	NH
EAST Search	08/26/2021	NH
NPL Search	08/26/2021	NH
PALM Inventor Search	03/25/2022	NH
EAST Search	03/25/2022	NH
NPL Search	03/25/2022	NH
PALM Inventor Search	06/16/2022	NH
SEARCH	06/16/2022	NH
NPL Search	06/16/2022	NH

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

Interference Search			
US Class/CPC Symbol	**US Subclass/CPC Group**	**Date**	**Examiner**
C07C	11/18, 2527/054, 2527/167, 2527/173, 2531/02, 65/21,	04/10/2021	NH
A61K	8/345, 2300/00, 2800/31, 8/60, 2800/412, 8/11, 8/732, 8/738, 31/197, 8/44, 8/73, 2800/48, 8/25, 8/34, 8/585, 8/92, 8/9789, 9/014, 2800/30, 31/198, 36/28, 36/534, 36/67	04/10/2021	NH
A61Q	15/00, 19/00, 13/00, 11/00, 5/00, 19/007, 19/08, 19/02, 17/005, 17/04, 19/10, 5/02, 7/00	04/10/2021	NH
C07C	11/18, 2527/054, 2527/167, 2527/173, 2531/02, 65/21,	06/16/2022	NH
A61K	8/345, 2300/00, 2800/31, 8/60, 2800/412, 8/11, 8/732, 8/738, 31/197, 8/44, 8/73, 2800/48, 8/25, 8/34, 8/585, 8/92, 8/9789, 9/014, 2800/30, 31/198, 36/28, 36/534, 36/67	06/16/2022	NH
A61Q	15/00, 19/00, 13/00, 11/00, 5/00, 19/007, 19/08, 19/02, 17/005, 17/04, 19/10, 5/02, 7/00	06/16/2022	NH

/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	Art Unit
	NANNETTE HOLLOMAN	1612

CPC								
Symbol							Type	Version
C07C	/	29	/	04			F	2013-01-01
C07C	/	31	/	20			I	2013-01-01
C07C	/	29	/	80			I	2013-01-01
B01J	/	23	/	06			I	2013-01-01
B01J	/	37	/	08			I	2013-01-01
B01J	/	21	/	04			I	2013-01-01
C11D	/	3	/	2044			I	2013-01-01
C07C	/	1	/	24			I	2013-01-01
C07C	/	29	/	48			I	2013-01-01
C07C	/	11	/	02			I	2013-01-01
C07C	/	11	/	107			I	2013-01-01
B01J	/	2523	/	27			A	2013-01-01
C07B	/	41	/	02			A	2013-01-01
C07B	/	63	/	00			A	2013-01-01
B01J	/	2523	/	31			A	2013-01-01
A61L	/	2	/	18			A	2013-01-01
A01N	/	31	/	02			A	2013-01-01
A61K	/	8	/	345			A	2013-01-01
A61Q	/	5	/	00			A	2013-01-01
A61Q	/	11	/	00			A	2013-01-01
A61Q	/	19	/	00			A	2013-01-01
C11D	/	3	/	48			A	2013-01-01
C07C	/	2521	/	04			A	2013-01-01

CPC Combination Sets									
Symbol							Type	Set	Ranking	Version
C07C	/	1	/	24			I	1	1	2013-01-01
C07C	/	11	/	02			I	1	2	2013-01-01
C07C	/	29	/	48			I	2	1	2013-01-01
C07C	/	31	/	20			I	2	2	2013-01-01

NONE		Total Claims Allowed:	
(Assistant Examiner)	(Date)	23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	11 July 2022	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	NONE

U.S. Patent and Trademark Office — Part of Paper No.: 20220705

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	**Art Unit**
	NANNETTE HOLLOMAN	1612

CPC Combination Sets								
Symbol					**Type**	**Set**	**Ranking**	**Version**
C07C	/	29	/	80	I	3	1	2013-01-01
C07C	/	31	/	20	I	3	2	2013-01-01

NONE		**Total Claims Allowed:**	
(Assistant Examiner)	(Date)	23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612	11 July 2022	O.G. Print Claim(s)	O.G. Print Figure
(Primary Examiner)	(Date)	1	NONE

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	**Art Unit**
	NANNETTE HOLLOMAN	1612

INTERNATIONAL CLASSIFICATION					
CLAIMED					
C07C29/04	/	29	/	04	

NON-CLAIMED				
	/		/	

US ORIGINAL CLASSIFICATION	
CLASS	**SUBCLASS**

CROSS REFERENCES(S)						
CLASS	**SUBCLASS (ONE SUBCLASS PER BLOCK)**					

NONE (Assistant Examiner) (Date)	**Total Claims Allowed:** 23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612 (Primary Examiner) 11 July 2022 (Date)	O.G. Print Claim(s) 1	O.G. Print Figure NONE

Issue Classification	Application/Control No.	Applicant(s)/Patent Under Reexamination
	16/262,902	Burgo et al.
	Examiner	**Art Unit**
	NANNETTE HOLLOMAN	1612

☐ Claims renumbered in the same order as presented by applicant ☐ CPA ☐ T.D. ☐ R.1.47

CLAIMS

Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original	Final	Original
	1		15		29		43		57		71		85	11	99
	2		16		30		44		58		72		86	12	100
	3		17		31		45		59		73		87	13	101
	4		18		32		46		60		74	1	88	14	102
	5		19		33		47		61		75	2	89	15	103
	6		20		34		48		62		76	3	90	16	104
	7		21		35		49		63		77	4	91	17	105
	8		22		36		50		64		78	5	92	18	106
	9		23		37		51		65		79	6	93	19	107
	10		24		38		52		66		80		94	20	108
	11		25		39		53		67		81	7	95	21	109
	12		26		40		54		68		82	8	96	22	110
	13		27		41		55		69		83	9	97	23	111
	14		28		42		56		70		84	10	98		

NONE (Assistant Examiner)	(Date)	**Total Claims Allowed:** 23	
/NANNETTE HOLLOMAN/ Primary Examiner, Art Unit 1612 (Primary Examiner)	11 July 2022 (Date)	O.G. Print Claim(s) 1	O.G. Print Figure NONE

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
P.O. Box 1450
Alexandria, Virginia 22313-1450
www.uspto.gov

APPLICATION NO.	ISSUE DATE	PATENT NO.	ATTORNEY DOCKET NO.	CONFIRMATION NO.
16/262,902	08/16/2022	11414365	INLX.0198U2/00484799	1028

171052 7590 07/27/2022

Cozen O'Connor - Inolex
1200 19th St. NW
Washington, DC 20036

ISSUE NOTIFICATION

The projected patent number and issue date are specified above.

Determination of Patent Term Adjustment under 35 U.S.C. 154 (b)
(application filed on or after May 29, 2000)

The Patent Term Adjustment is 7 day(s). Any patent to issue from the above-identified application will include an indication of the adjustment on the front page.

If a Continued Prosecution Application (CPA) was filed in the above-identified application, the filing date that determines Patent Term Adjustment is the filing date of the most recent CPA.

Applicant will be able to obtain more detailed information by accessing the Patent Application Information Retrieval (PAIR) WEB site (http://pair.uspto.gov).

Any questions regarding the Patent Term Extension or Adjustment determination should be directed to the Office of Patent Legal Administration at (571)-272-7702. Questions relating to issue and publication fee payments should be directed to the Application Assistance Unit (AAU) of the Office of Patents Stakeholder Experience (OPSE), Stakeholder Support Division (SSD) at (571)-272-4200.

INVENTOR(s) (Please see PAIR WEB site http://pair.uspto.gov for additional inventors):

Rocco V. Burgo, Mullica Hill, NJ;
Michael E. Wright, Hurricane, UT;
Gary B. Mosser, Tabernacle, NJ;
Michael J. Fevola, Belle Mead, NJ;

APPLICANT(s) (Please see PAIR WEB site http://pair.uspto.gov for additional applicants):

Inolex Investment Corporation, Wilmington, DE

The United States represents the largest, most dynamic marketplace in the world and is an unparalleled location for business investment, innovation, and commercialization of new technologies. The USA offers tremendous resources and advantages for those who invest and manufacture goods here. Through SelectUSA, our nation works to encourage and facilitate business investment. To learn more about why the USA is the best country in the world to develop technology, manufacture products, and grow your business, visit SelectUSA.gov.