18304 U.S. PTO
020606

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UTILITY PATENT APPLICATION TRANSMITTAL

(Only for new nonprovisional applications under 37 CFR 1.53(b))

Attorney Docket No.	FDS-029388-US
First Inventor	CLEVELAND, et al.
Title	BIODEGARADABLE PAPER-BASED ...
Express Mail Label No.	EQ239461176US

112387 U.S. PTO
11/348150
020606

APPLICATION ELEMENTS

See MPEP chapter 600 concerning utility patent application contents.

1. ☑ **Fee Transmittal Form** (e.g., PTO/SB/17)
   (Submit an original and a duplicate for fee processing)
2. ☐ **Applicant claims small entity status.**
   See 37 CFR 1.27.
3. ☑ **Specification** [*Total Pages* 28]
   Both the claims and abstract must start on a new page
   (For information on the preferred arrangement, see MPEP 608.01(a))
4. ☑ **Drawing(s)** (*35 U.S.C. 113*) [*Total Sheets* 5]
5. **Oath or Declaration** [*Total Sheets* _____]
   a. ☐ Newly executed (original or copy)
   b. ☐ A copy from a prior application (37 CFR 1.63(d))
   (for continuation/divisional with Box 18 completed)
   i. ☐ DELETION OF INVENTOR(S)
   Signed statement attached deleting inventor(s) name in the prior application, see 37 CFR 1.63(d)(2) and 1.33(b).
6. ☑ **Application Data Sheet.** See 37 CFR 1.76
7. ☐ **CD-ROM or CD-R** in duplicate, large table or Computer Program *(Appendix)*
   ☐ Landscape Table on CD
8. **Nucleotide and/or Amino Acid Sequence Submission**
   (if applicable, items a. – c. are required)
   a. ☐ Computer Readable Form (CRF)
   b. Specification Sequence Listing on:
      i. ☐ CD-ROM or CD-R (2 copies); or
      ii. ☐ Paper
   c. ☐ Statements verifying identity of above copies

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ACCOMPANYING APPLICATION PARTS

9. ☐ **Assignment Papers** (cover sheet & document(s))
   Name of Assignee _____
10. ☐ **37 CFR 3.73(b) Statement** *(when there is an assignee)* ☐ **Power of Attorney**
11. ☐ **English Translation Document** *(if applicable)*
12. ☐ **Information Disclosure Statement** (PTO/SB/08 or PTO-1449)
    ☐ Copies of citations attached
13. ☐ **Preliminary Amendment**
14. ☑ **Return Receipt Postcard** (MPEP 503)
    (Should be specifically itemized)
15. ☐ **Certified Copy of Priority Document(s)**
    (if foreign priority is claimed)
16. ☐ **Nonpublication Request** under 35 U.S.C. 122(b)(2)(B)(i).
    Applicant must attach form PTO/SB/35 or equivalent.
17. ☑ **Other:** cover letter

18. If a CONTINUING APPLICATION, *check appropriate box, and supply the requisite information below and in the first sentence of the specification following the title, or in an Application Data Sheet under 37 CFR 1.76:*

☐ Continuation ☐ Divisional ☐ Continuation-in-part (CIP) of prior application No.: .....

Prior application information: *Examiner* _____ *Art Unit:* _____

19. CORRESPONDENCE ADDRESS

☑ The address associated with Customer Number: 01726 *OR* ☐ Correspondence address below

Name					
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City		State		Zip Code	
Country		Telephone		Email	

Signature	Melvin D. Fletcher	Date	FEBRUARY 6, 2006
Name (Print/Type)	MELVIN D. FLETCHER	Registration No. (Attorney/Agent)	36,137

This collection of information is required by 37 CFR 1.53(b). 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. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

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Effective on 12/08/2004.
Fees pursuant to the Consolidated Appropriations Act, 2005 (H.R. 4818).

FEE TRANSMITTAL
For FY 2005

☐ Applicant claims small entity status. See 37 CFR 1.27

TOTAL AMOUNT OF PAYMENT	($)	5650.00

Complete if Known	
Application Number	NEW APPLICATION
Filing Date	FEBRUARY 6, 2006
First Named Inventor	CLEVELAND, et al.
Examiner Name	
Art Unit	
Attorney Docket No.	FDS-029388-US

METHOD OF PAYMENT (check all that apply)

☐ Check ☐ Credit Card ☐ Money Order ☐ None ☐ Other (please identify): _____

☑ Deposit Account Deposit Account Number: 09-0525 Deposit Account Name: INTL PAPER CO

For the above-identified deposit account, the Director is hereby authorized to: (check all that apply)

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FEE CALCULATION

1. BASIC FILING, SEARCH, AND EXAMINATION FEES

	FILING FEES		SEARCH FEES		EXAMINATION FEES		
Application Type	Fee ($)	Small Entity Fee ($)	Fee ($)	Small Entity Fee ($)	Fee ($)	Small Entity Fee ($)	Fees Paid ($)
Utility	300	150	500	250	200	100	1000.00
Design	200	100	100	50	130	65	
Plant	200	100	300	150	160	80	
Reissue	300	150	500	250	600	300	
Provisional	200	100	0	0	0	0	

2. EXCESS CLAIM FEES

Fee Description	Fee ($)	Small Entity Fee ($)
Each claim over 20 (including Reissues)	50	25
Each independent claim over 3 (including Reissues)	200	100
Multiple dependent claims	360	180

Total Claims		Extra Claims	Fee ($)	Fee Paid ($)
73	- 20 or HP =	53	x 50.00	= 2650.00

HP = highest number of total claims paid for, if greater than 20.

Indep. Claims		Extra Claims	Fee ($)	Fee Paid ($)
13	- 3 or HP =	10	x 200.00	= 2000.00

HP = highest number of independent claims paid for, if greater than 3.

Multiple Dependent Claims	
Fee ($)	Fee Paid ($)

3. APPLICATION SIZE FEE

If the specification and drawings exceed 100 sheets of paper (excluding electronically filed sequence or computer listings under 37 CFR 1.52(e)), the application size fee due is $250 ($125 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).

Total Sheets		Extra Sheets		Number of each additional 50 or fraction thereof	Fee ($)	Fee Paid ($)
	- 100 =		/ 50 =	(round up to a whole number) x		=

4. OTHER FEE(S)

	Fees Paid ($)
Non-English Specification, $130 fee (no small entity discount)	
Other (e.g., late filing surcharge):	

SUBMITTED BY

Signature	Melvin D. Fletcher	Registration No. (Attorney/Agent) 36,137	Telephone 513/248-6069
Name (Print/Type)	MELVIN D. FLETCHER		Date 2/6/2006

This collection of information is required by 37 CFR 1.136. 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, 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.**

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INTERNATIONAL PAPER

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LABEL NO. EQ239461176US

February 6, 2006

Commissioner for Patents
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RE: SUBMISSION OF NON-PROVISIONAL APPLICATION
Applicant : CLEVELAND, et al.
Title : BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN.....
Our Ref. : FDS-029388-US

Dear Commissioner:

Enclosed herewith for filing, Applicant(s) respectfully submit(s) the following checked items:

- [x] Utility Patent Application Transmittal, Form PTO/SB/05, 1 sheet(s);
- [x] Fee Transmittal, Form PTO/SB/17, 1 sheet(s);
- [x] Application Data Sheet, 3 sheet(s);
- [x] Patent Specification, 28 sheet(s);
- [x] 5 Figures arranged on 5 sheet(s);
- [ ] Information Disclosure Statement, Form PTO/SB/08, _____ sheet(s), and one copy of each reference cited thereon;
- [ ] Signed Inventors Declaration(s), _____ sheet(s);
- [ ] Nonpublication Request, Form PTO/SB/35, _____ sheet(s); and,
- [x] a postage-prepaid, return-receipt postcard with a listing of the above-checked items.

Please stamp the enclosed postcard and return same to me to indicate your receipt of the above-listed items. Please feel free to contact me if you have any questions concerning the above or the enclosed.

With kindest regards,

Melvin D. Fletcher
Attorney for Applicant(s)

Enclosures
MDF/ jat

BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOISTURE BARRIER PROPERTIES AND METHOD FOR MAKING BIODEGRADABLE PAPER-BASED LAMINATE

Background of the Invention:

Field Of Invention:

[0001] This invention relates to biodegradable paper-based laminates and methods for making biodegradable paper-based laminates.

Description of the Related Art:

[0002] Paper-based laminates commonly include a substrate of paperboard, container board, or other form of paper. Due to paper's inherent limitations relative to its porosity to deleterious liquids or gases, its physical properties such as tear resistance, and other limitations for packaging uses, the paper-based substrate is coated or laminated with one or more layers of polymeric materials, which are selected to obviate one or more of the limitations of the paper-board as a packaging material. These polymeric materials such as, but not limited to, low-density polyethylene (LDPE), nylon, polyethylene terephthalate (PET), and ethylene vinyl alcohol copolymer (EVOH) are not generally considered biodegradable. Paper-based laminates are useful in the formation of packaging for a large variety of products. Such packaging is commonly used once or a minimal number of times and then disposed. The volume of usage of such laminates has created potential problems with respect to their biodegradability in landfills, etc. Uses of paper-based laminates include packaging for food products (liquids, solids, hot or cold), ream wrap, sandwich wrap, and a myriad of other uses.

[0003] Paper-based laminates for food service uses are typically extrusion coated with low-density polyethylene (LDPE) or other similar polymer(s) in order to hold liquids for a longer period of time without leaking or becoming soft as is common with 100% paper cups. For example, cups for hot beverages such as coffee generally have a layer of LDPE on the inside

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for liquid resistance and sealing. Cold drink cups for soft drinks and the like are typically coated with LDPE on both sides to prevent condensation that forms on the outside of the cup from softening the paper. LDPE coat weights of 0.5 - 1.5 mils (7.2 - 21.6 lb/3000 ft^2) are common.

[0004] Many paper-based laminates are used in disposable packaging once or a very minimal number of times then disposed. The LDPE coating is not readily biodegradable, and therefore, the cup might remain in a landfill for many years without degrading. The use of one or more biodegradable polymers instead of LDPE is desirable to render the used cups more "environmentally friendly".

[0005] Paper-based laminates for food service uses are typically extrusion coated with low-density polyethylene (LDPE) or other similar polymers in order to hold liquids for a longer period of time without leaking or becoming soft, as is common with 100% paper cups. For example, cups for hot beverages such as coffee generally have a layer of LDPE on the inside for liquid resistance and sealing. Cold drink cups for soft drinks and the like are typically coated with LDPE on both sides to prevent condensation that forms on the outside of the cup from softening the paper. LDPE coat weight of 0.5 – 1.5 mils (7.2 -21.6 lb/ 3000 ft^2) are common.

[0006] In addition to cups, other coated paper products such as gable-top cartons, folding cartons, corrugated boxes, paper pouches, sandwich wraps, and ream wrap may also benefit from including an environmentally friendly biodegradable material.

[0007] Renewable-resource-based polymers exist. These polymers produced from chemicals that can be grown. This allows for greater supply and price stability.

[0008] In addition, these materials are biodegradable and compostable. In Europe and Asia, where landfill space is very limited, compostable renewable-resource-based materials are

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preferred. In additional, municipal composting is a growing trend in North America, especially where landfills are filling too quickly and related costs are rising rapidly.

[0009] Compostable renewable-resource-based materials include polylactic acid (PLA) and polyhydroxyalkanoates (PHA).

[0010] Usually, "green" materials like PLA and PHA are difficult to process via extrusion because of their sensitivity to moisture and high temperatures.

Summary of Invention:

[0011] It is accordingly an object of the invention to provide a biodegradable paper-based laminate with oxygen and moisture barrier properties that overcomes the hereinafore-mentioned disadvantages of the heretofore-known devices of this general type and that is fully biodegradable/compostable, paper-based, and suitable for use in packaging of various types.

[0012] With the foregoing and other objects in view, there is provided, in accordance with the invention, a paper-based substrate having one or more layers of material applied thereto. Such layers each are fully biodegradable/compostable and each layer of material contributes selected desirable properties, either alone or in combination, with one or more additional layers. The sum of the layers is a fully biodegradable/compostable material having all of those properties that are desired or required to render the laminate useful in the packaging of a given product.

[0013] Specifically, in accordance with one aspect of the present invention, there is provided a multi-layered paper-based laminate having oxygen and/or water vapor barrier properties, wherein the laminate is fully biodegradable and compostable. In many instances, the individual layer components of the laminate of the present invention, when applied singly to a substrate fail to provide the desired properties for the laminate. Rather, only when these layers are coextruded onto an underlying layer or onto the substrate itself do the layers contribute the desired properties to the laminate.

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[0014] With the objects of the invention in view, there is also provided a method for making biodegradable paper products with barrier properties. The method includes providing a substrate having a surface and extruding a barrier layer onto the surface of the substrate. The barrier layer includes a degradable material such as polyvinyl alcohol, polylactic acid or polyhydroxyalkonoate. The coatweight of the degradable material is within a range from 8 to 20 pounds per three-thousand square feet. A second barrier layer can be extruded on an opposing side of the substrate.

[0015] To promote adhesion of the extruded polymer, the method can include applying primer to the substrate before the extruding step. Preferably, an aqueous primer is used. Water is removed from the aqueous primer immediately prior the extruding step. The primer is preferably a starch emulsion, a soy protein emulsion, polyvinyl acetate, or polyvinyl alcohol. The coatweight of the primer is kept within a range from 0.5 to 5 pounds per three-thousand square feet.

[0016] To reduce the high cost of PLA and PHA relative LDPE and PP, an inexpensive filler can be added to the degradable material before the extruding step. The filler can be calcium carbonate, talc, diatomaceous earth, or clay. An amount of the filler ranging from 5 to 20 percent by weight in the degradable material has been found to efficacious.

[0017] When polyhydroxyalkanoate is the degradable material, the extruded polymer may block. Because the glass transient temperature (T_g) of PHA is usually below room temperature, PHA remains soft and continues to crystallize for period after processing. As a coated roll sets following extrusion, the material remains tacky, which can lead to layers sticking together or "blocking". Accordingly, an antiblock material can be added to the barrier layer after the extruding step. Suitable antiblock materials include erucamide, calcium carbonate, and talc. An amount of the antiblock material in a range from 0.1 to 5 percent by weight has been found to be efficacious.

[0018] Other features that are considered as characteristic for the invention are set forth in the appended claims.

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[0019] Although the invention is illustrated and described herein as embodied in a biodegradable paper-based laminate with oxygen and moisture barrier properties and a method for making the laminates, it is nevertheless not intended to be limited to the details shown, since various modifications and structural changes may be made therein without departing from the spirit of the invention and within the scope and range of equivalents of the claims.

[0020] The construction and method of formation of the invention, however, together with additional objects and advantages thereof will be best understood from the following description of specific embodiments when read in connection with the accompanying drawings.

Brief Description of the Drawings:

[0021] Fig. 1 is a schematic representation of a laminate exhibiting oxygen barrier properties according to the present invention.

[0022] Fig. 2 is a schematic representation of a laminate utilizing degradable polyolefins and degradable tie layers to adhere the degradable polyolefins to the oxygen barrier layer.

[0023] Figs. 3A-3C are schematic representations of three embodiments of laminates shown generally in Fig. 1.

[0024] Figs. 4A-4C are schematic representations of three embodiments of laminates that combine the laminates shown in Figs. 1-2.

[0025] Fig. 6 is a schematic representation of a laminate exhibiting liquid and vapor water barrier properties according to the present invention and having one barrier layer.

[0026] Fig. 7 is a schematic representation of a laminate exhibiting liquid and vapor water barrier properties according to the present invention and having two barrier layers.

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Detailed Description Of The Preferred Embodiments:

[0027] For purposes of the present disclosure, the laminates of the present invention are divided into two groups: (1) oxygen barriers and (2) liquid and vapor water barriers.

Oxygen Barriers:

[0028] A laminate according to the present invention that exhibits oxygen barrier properties is generally depicted in Fig. 1.

[0029] As seen in Fig. 1, the depicted substrate is paperboard, such as bleached or unbleached (natural) kraft sheet or bleached or unbleached (natural) paperboard.

[0030] Layer A, as depicted, is to be disposed on the outside of the package formed from the laminate. Therefore, this layer may include a polymer that will provide the substrate adequate protection from liquid and vapor water in the surrounding environment. It will also act as the print medium. This material may be degradable polyolefins (dPE) or polyhydroxyalkanoates (PHA) with a film coat weight of 5-20 lb / 3000 ft^2. Polylactic acid (PLA) and other biodegradable polyesters may also be used in the same coat weight range, but offer less water vapor barrier than dPE or PHA.

[0031] A degradable polyolefin (dPE) is a blend of a transition metal carboxylate catalyst with polyethylene and polypropylene. Examples of transition metals used in the catalyst may include: iron, cadmium, cobalt, and manganese. Two commercial suppliers of this additive are Environmental Plastics, Inc. (EPI) and Willow Ridge Plastics, Inc.

[0032] Suitable biodegradable polyesters are sold under the trade names BASF ECOFLEX, EASTMAN EASTAR BIO, and DUPONT BIOMAX. These materials degrade by a combination of hydrolysis and biodegradation (consumption by microorganisms).

[0033] Layer B is chosen from materials that will coat the substrate to protect the following layers from moisture from the substrate and the exterior environment. This material may

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include a 5-20 lb / 3000 ft^2 coating of dPE, PHA, or PLA. Alternatively, this layer may provide an oxygen barrier and is formed from a 5-20 lb / 3000 ft^2 coating of PVOH, biodegradable polyester, PHA, or PLA.

[0034] Layer C generally includes a material that will provide the oxygen barrier. Suitable materials include a 2-15 lb / 3000 ft^2 layer of polyvinyl alcohol (PVOH), which can be applied as an aqueous or extruded coating. However, this layer may not be necessary if Layer B includes an oxygen barrier material, or this layer may include a degradable tie layer. In the latter instance, this would include a degradable material (dTie), which is a blend of a transition metal carboxylate catalyst additive described previously with a maleic anhydride functionalized low-density polyethylene (LDPE) or linear low-density polyethylene (LLDPE) (such as those sold under the trade name PLEXAR® from Equistar Chemical). A suitable layer thickness for a dTie layer is generally 2-5 lb / 3000 ft^2.

[0035] Layer D is the product contact layer. As such it provides resistance to penetration by vapor (water), liquid (i.e. water, oil, etc.), and solid materials and aids in sealing the package. This layer preferably has a coat weight of 5-20 lb / 3000 ft^2 and includes dPE or PHA, with PLA and other biodegradable polyesters being useable in certain situations.

[0036] If dPE is used in Layers B and D, then a dTie layer may need to be applied between Layers B and C and between Layers C and D to provide adequate adhesion thereby creating a five layer structure on the matte side, as depicted in Fig. 2.

[0037] The substrate of the embodiment depicted in Fig. 2 may be bleached or unbleached (natural) kraft sheet or bleached or unbleached (natural) paperboard.

[0038] Layer E of this embodiment is located on the outside of the package and overlying one flat surface of the substrate. Thus, Layer E will include a polymer that can provide the substrate adequate protection from liquid and vapor water in the surrounding environment. Layer E will also act as the print medium. This material may be dPE or PHA applied at a film

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coat weight of 5-20 lb / 3000 ft^2. PLA and other degradable polyesters may also be used in the same coat weight range, but offer less water vapor barrier than dPE or PHA.

[0039] Layers F and J act as moisture resistant layers to protect the oxygen barrier layer(s). These layers can be dPE, PHA, or other degradable polyesters in coat weights of 5-20 lb / 3000 ft^2. Layer H preferably includes the primary oxygen barrier material of the laminate. Suitable materials include a 2-15 lb / 3000 ft^2 layer of PVOH, PHA, or PLA, which can be applied as an aqueous or extruded coating. In many instances, layers G and I then include degradable tie (dTie) to ensure adhesion between layers F and H and layers H and J. A suitable thickness for a degradable tie layer is 2-5 lb / 3000 ft^2.

[0040] Alternatively, both Layers G and I can include the primary oxygen barrier materials of the laminate. Suitable materials include a 2-15 lb / 3000 ft^2 layer of polyvinyl alcohol (PVOH), PHA, PLA, or other biodegradable polyesters, which can be applied as an aqueous or extruded coating.

[0041] If dPE of a layer thickness of 5-20 lb / 3000 ft^2 is used in layers F, H, and J, dTie layers generally of thickness 2-5 lb / 3000 ft^2 will be needed between is Layers F and G, G and H, H and I, and I and J to provide adequate adhesion.

[0042] Combination of Figs. 1 and 2 should also be considered to further enhance the overall functionality of the laminate. For example, the three layer laminate of Fig. 1 may be applied to the substrate as a single coextrusion step, followed by a five to nine layer laminate of Fig. 2 as another single coextrusion step, provided that adequate adhesion is attained between the coextrusions.

[0043] The laminates of Figs. 1 and 2 are particularly advantageous for packaging products such as specialty milks, fruit juices, juice drinks, and other beverages in gable top cartons, pouches, and the like where a shelf life of several weeks to several months is required. The

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oxygen barrier layer(s) prevent oxidation of the liquid product, thereby aiding in the retention of vitamins, aroma/flavor, and product color.

[0044] The laminates shown in Figs. 3A-3C are formed from biodegradable materials as defined in the text and in Fig. 1. Figs. 3A-3C are not meant to be limiting and other combinations of the materials described may also provide suitable laminates.

[0045] The laminates shown in Figs. 4A-4C are formed from biodegradable materials as defined in the text and via the combination of Figs. 1 and 2. All of the laminates in Figs. 4A-4C are expected to have improved oxygen barrier performance in comparison to the laminates in Figs. 3A-3C. Once again, Figs. 4A-4C are not meant to be limiting and other combinations of the materials described may also provide suitable laminates.

[0046] The laminates shown in Figs. 5A-5C are formed from biodegradable materials as defined in the text and via the combination of Figs. 1 and 2. All of the laminates in Figs. 5A-5C are expected to have improved oxygen barrier performance in comparison to the laminates in Figs. 4A-4C. Once again, Figs. 5A-5C are not meant to be limiting and other combinations of the materials described may also provide suitable laminates.

Liquid And Vapor Water Barriers:

[0047] Figs. 6 and 7 depict embodiments of the present laminate, which is are useful for providing moisture (vapor and liquid) barrier structures according to the present invention.

[0048] As depicted in Fig. 6, the substrate may include bleached or unbleached (natural) kraft sheet or bleached or unbleached (natural) paperboard. This substrate is coated on one (product contact) side for liquid retention and is a candidate for constructing drinking cups for hot liquids. Layer K has a thickness of 5-30 lb / 3000 ft^2 may be dPE, PHA, PLA, or other degradable polyesters.

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[0049] In the structure depicted in Fig. 7, a paperboard substrate (bleached or unbleached (natural) kraft sheet or bleached or unbleached (natural) paperboard) is coated on both sides for packaging cold liquids or solids that are moisture sensitive. Degradable polyesters, such as dPE, PHA, PLA, may be used to construct Layers L and M. Suitable layer thicknesses for Layers L and M are 5-30 lb / 3000 ft^2.

[0050] In addition to cups for hot and cold liquids, the laminates of Figs. 6 and 7 may also be utilized in gable top cartons or pouches for the storage of liquid and dry products that do not require an oxygen barrier. An example is fresh, pasteurized milk.

[0051] As desired, calcium carbonate may be added to one or more of the individual layers of the extrusion or aqueous coatings in Figs. 1, 2, 6, and 7 as a cost savings measure and to provide an increase in the degradation rate by displacement of some of the biodegradable resin material. Other possible organic and inorganic fillers may be employed with, or in lieu of, calcium carbonate, including starch, clay, kaolin, talc, cellulose fibers, and diatomaceous earth.

[0052] In some instances at processing temperatures typically used for extrusion coating applications, a high rate of degradation can occur which creates low molecular weight products and a potentially unpleasant off-odor. To slow this reaction, vitamin E, tetrakis-(methylene-(3,5-di-terbutyl-4-hydrocinnamate)methane (which is sold under the trade name IRGANOX 1010), and/or other antioxidants can be added at loadings from a few parts-per-million up to about five weight percent (<5%). In addition, zeolites (or molecular sieves) can be used to scavenge all or some of the low molecular weight products thereby reducing unwanted odors. Zeolites are also typically added at loadings up to about five weight percent (<5%).

[0053] Furthermore, dPE is a technology which involves using degradation-promoting additives in polyolefins such as polyethylene. These additives involve a mixture of polymer, filler, and metal carboxylate catalyst. In one embodiment, the inventors combine dPE with a calcium carbonate additive to disrupt the polymer matrix and reduce the overall amount of polymer present in a given layer(s), thereby affecting both enhanced biodegradation and cost savings.

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[0054] In addition, d-tie is a development of the present inventors involving the addition of a metal catalyst to a tie resin (i.e., ethylene-co-maleic anhydride) to produce a degradable tie layer product.

[0055] A preferred method of forming laminates that include the barrier layers utilizes extrusion coating. The method can be applied in both single-side (1-S) coated and double-side (2-S) coated. The desired coatweights of PLA or PHA are 8-20 pounds per three-thousand square feet.

[0056] To promote adhesion of the extruded PHA and PLA polymers, a primer is included. The primer is applied and the water removed prior to an extruder station. With the improved adhesion, one can achieve a lower extrude coatweight and increased linespeed. Examples of priming materials include aqueous solutions or emulsions of starch, soy protein, polyvinyl acetate, and polyvinyl alcohol. Typical coat weights of the primer are in the range of 0.5 – 5 pounds per three-thousand square feet.

Additives and fillers can be used to reduce the relative cost of PHA and PLA. Preferred additives include calcium carbonate, talc, diatomaceous earth, and clay. The preferred amount of these fillers is usually 5-20% by weight.

In the case of PHA, its glass transition temperate (T_g) is usually below room temperature. Preferred antiblock materials include erucamide, calcium carbonate, and talc. These materials are blended to 0.1-5% by weight and "bloom" to the surface. This creates a microscopic roughness that hinders blocking while the polymer continues to crystallize.

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We claim:

1. A laminate with oxygen barrier properties, comprising:

a substrate having an outside surface and an inside surface;

a polymeric protectant layer disposed on said outside surface of said substrate for protecting said substrate from liquid and vapor water surrounding the laminate;

a moisture protective layer disposed on said inside surface of said substrate for blocking moisture from said substrate and an exterior environment;

one of an oxygen barrier layer and a degradable tie layer being disposed on said moisture protective layer, said oxygen barrier layer being present when said moisture protective layer is not an oxygen barrier, said degradable tie layer for increasing adhesion and being present when said moisture protective layer is an oxygen barrier; and

a product contact layer for preventing penetration by vapor, liquid, and solid contents disposed on said oxygen barrier layer or said degradable tie layer.

2. The laminate according to claim 1, wherein said substrate is paperboard.

3. The laminate according to claim 2, wherein said paperboard is selected from the group consisting of bleached kraft sheet, unbleached kraft sheet, bleached paperboard, and unbleached paperboard.

4. The laminate according to claim 1, wherein said polymeric protectant layer acts as a print medium.

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5. The laminate according to claim 1, wherein said polymeric protectant layer is selected from the group consisting of a degradable polyolefin, a polyhydroxyalkanoate, polylactic acid, and a biodegradable polyester.

6. The laminate according to claim 5, wherein said polymeric protectant layer has a film coat weight from five to twenty pounds per three-thousand square feet.

7. The laminate according to claim 5, wherein said degradable polyolefin is a blend of a transition metal carboxylate catalyst and a polyolefin.

8. The laminate according to claim 7, wherein said polyolefin is selected from the group consisting of polyethylene and polypropylene.

9. The laminate according to claim 7, wherein said transitional metal carboxylate catalyst includes a metal selected from the group consisting of iron, cadmium, cobalt, and manganese.

10. The laminate according to claim 1, wherein said moisture protective layer is selected from the group consisting of a degradable polyolefin, a polyhydroxyalkanoate, and polylactic acid.

11. The laminate according to claim 10, wherein said moisture protective layer has a film coat weight from five to twenty pounds per three thousand square feet.

12. The laminate according to claim 1, wherein said moisture protective layer is an oxygen barrier.

13. The laminate according to claim 12, wherein said oxygen barrier is formed from a polymer selected from the group consisting of polyvinyl alcohol, a birdegradable polyester, a polyhydroxyalkanoate, and polylactic acid.

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14. The laminate according to claim 12, wherein said oxygen barrier has a film coat weight from five to twenty pounds per three-thousand square feet.

15. The laminate according to claim 1, including said oxygen barrier layer, said oxygen barrier layer including polyvinyl alcohol.

16. The laminate according to claim 15, wherein said polyvinyl alcohol has a film coat weight from two to fifteen pounds per three-thousand square feet.

17. The laminate according to claim 1, including said degradable tie material, said degradable tie material including a transitional metal carboxylate catalyst and a maleic anhydride functionalized polyethylene.

18. The laminate according to claim 17, wherein said degradable polyethylene is selected from the group consisting of maleic anhydride functionalized polypropylene, high-density polyethylene, low-density polyethylene and linear low-density polyethylene.

19. The laminate according to claim 17, wherein said transition metal carboxylate catalyst includes a metal selected from the group consisting of iron, cadmium, cobalt, and manganese.

20. The laminate according to claim 1, including said degradable tie layer, said degradable tie layer having a film coat weight from two to five pounds per three-thousand square feet.

21. The laminate according to claim 1, wherein said product contact layer has a coat weight of five to twenty pounds per three thousand square feet.

22. The laminate according to claim 1, wherein said product contact layer includes a polymer selected from the group consisting of a degradable polyolefin, a polyhydroxyalkanoate, polylactic acid, and a biodegradable polyester.

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23. The laminate according to claim 1, including said oxygen barrier layer; and

at least one of said moisture protective layer and said product contact layer is a dPE layer including a degradable polyolefin; and

a dTie layer is disposed between said dPE layer and said oxygen barrier layer for increasing adhesion between said dPE layer and said oxygen barrier layer.

24. The laminate according to claim 23, wherein:

said moisture protective layer and said product contact layer are both dPE layers including a degradable polyolefin;

a first dTie layer is disposed between said product contact layer and said oxygen barrier layer; and

a second dTie layer is disposed between said moisture protective layer and said oxygen barrier layer.

25. The laminate according to claim 23, wherein said dPE layer includes a transition metal catalyst and a polyolefin.

26. The laminate according to claim 25, wherein said polyolefin is selected from the group consisting of polyethylene, polypropylene, and polystyrene.

27. The laminate according to claim 25, wherein said transition metal catalyst is selected from the group consisting of iron, cadmium, cobalt, and manganese.

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28. The laminate according to claim 23, wherein said moisture protective layer and said product contact layer include a degradable polymer selected from the group consisting of dPE and PHA.

29. The laminate according to claim 23, wherein said moisture protective layer and said product contact layer each have a coat weight from five to twenty pounds per three-thousand square feet.

30. The laminate according to claim 23, wherein said oxygen barrier layer is selected from the group consisting of polyvinyl alcohol, a polyhydroxyalkanoate, and polylactic acid.

31. The laminate according to claim 30, wherein said oxygen barrier layer has a coating weight from two to fifteen pounds per three-thousand square feet.

32. A laminate with oxygen barrier properties, comprising:

a substrate having an outside surface and an inside surface;

a first degradable polyolefin layer disposed on said outside surface of said substrate;

a moisture protectant layer disposed on said inside surface of said substrate including a biodegradable polymer selected from the group consisting of polyester, polyvinyl alcohol, polyhydroxyalkanoate, and polylactic acid;

a degradable tie layer disposed on said moisture protectant layer; and

a second degradable polyolefin layer disposed on said degradable tie layer.

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33. A laminate with oxygen barrier properties, comprising:

a substrate having an outside surface and an inside surface;

a polymeric protectant layer disposed on said outside surface of said substrate and including a polymer selected from the group consisting of polyhydroxyalkanoate and polylactic acid;

a biodegradable polyester layer disposed on said inside surface of said substrate; and

a product contact layer for resisting penetration disposed on said biodegradable polyester layer and including a polymer selected from the group consisting of polyhydroxyalkanoate and polylactic acid.

34. A laminate with oxygen barrier properties, comprising:

a substrate having an outside surface and an inside surface;

a first biodegradable polyester layer disposed on said outside surface of said substrate;

a moisture protectant layer disposed on said inside surface of said substrate including a biodegradable polymer selected from the group consisting of a polyhydroxyalkanoate and polylactic acid;

a degradable tie layer disposed on said moisture protectant layer; and

a second biodegradable polyester layer disposed on said degradable tie layer.

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35. A laminate with oxygen barrier properties, comprising:

a substrate having an outside surface and an inside surface;

a polymeric protectant layer disposed on said outside surface of said substrate and including a polymer selected from the group consisting of polyhydroxyalkanoate and polylactic acid;

a polyvinyl alcohol layer disposed on said inside surface of said substrate; and

a product contact layer for resisting penetration disposed on said polyvinyl alcohol layer and including a polymer selected from the group consisting of polyhydroxyalkanoate and polylactic acid.

36. A laminate with oxygen barrier properties, comprising:

a substrate having an outside surface and an inside surface;

a first degradable polyolefin layer disposed on said outside surface of said substrate;

a moisture protectant layer disposed on said inside surface of said substrate and including a biodegradable polymer selected from the group consisting of polyhydroxyalkanoate, polylactic acid, and polyester;

a first degradable tie layer disposed on said moisture protectant layer;

a second degradable polyolefin layer disposed on said first degradable tie layer;

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a third degradable polyolefin layer disposed on said second degradable polyolefin layer;

a second degradable tie layer disposed on said third degradable polyolefin layer;

a polyvinyl alcohol layer disposed on said second degradable tie layer;

a third degradable tie layer disposed on said polyvinyl alcohol layer; and

a second degradable polyolefin layer disposed on said third degradable tie layer.

37. A laminate with oxygen barrier properties, comprising:

a substrate having an outside surface and an inside surface;

a first biodegradable polyester layer disposed on said outside surface of said substrate;

a first moisture protectant layer disposed on said inside surface of said substrate and including a polymer selected from the group consisting of a polyhydroxyalkanoate and polylactic acid;

a first degradable tie layer disposed on said first moisture protectant layer;

a second biodegradable polyester layer disposed on said first degradable tie layer;

a third biodegradable polyester layer disposed on said second biodegradable polyester layer;

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a second degradable tie layer disposed on said third biodegradable polyester layer;

a second oxygen barrier layer disposed on said second degradable tie layer and including a biodegradable polymer selected from the group consisting of polyvinyl alcohol, a polyhydroxyalkanoate, polylactic acid, and biodegradable polyester;

a third degradable tie layer disposed on said second moisture protectant layer; and

a fourth polyester layer disposed on said third degradable tie layer.

38. A laminate with oxygen barrier properties, comprising:

a substrate having an outside surface and an inside surface;

a polymeric protectant layer disposed on said outside surface of said substrate and including a polymer selected from the group consisting of polyhydroxyalkanoate and polylactic acid;

a first polyvinyl alcohol layer disposed on said inside surface of said substrate;

a moisture protectant layer disposed on said first polyvinyl alcohol layer and including a biodegradable polymer selected from the group consisting of a polyhydroxyalkanoate and polylactic acid;

a second polyvinyl alcohol layer disposed on said moisture protectant layer; and

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a product contact layer disposed on said second polyvinyl alcohol layer and including a polymer selected from the group consisting of polyhydroxyalkanoate and polylactic acid.

39. A laminate with oxygen barrier properties, comprising:

a substrate having an outside surface and an inside surface;

a first degradable polyolefin layer disposed on said surface of said substrate;

a polymeric protectant layer disposed on said inside surface of said substrate and including a polymer selected from the group consisting of polyhydroxyalkanoate and polylactic acid;

a first polyvinyl alcohol layer disposed on said polymeric protectant layer;

a first degradable tie layer disposed on said first polyvinyl alcohol layer;

a second degradable polyolefin layer disposed on said first degradable tie layer;

a second degradable tie layer disposed on said second degradable polyolefin layer;

a second polyvinyl alcohol layer disposed on said second degradable tie layer;

a third degradable tie layer disposed on said second polyvinyl alcohol layer; and

a third degradable polyolefin layer disposed on said third degradable tie layer.

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40. A laminate with oxygen barrier properties, comprising:

a substrate having an outside surface and an inside surface;

a first degradable polyolefin layer disposed on said outside surface of said substrate;

a polyvinyl alcohol layer disposed on said inside surface of said substrate;

a first degradable tie layer disposed on said polyvinyl alcohol layer;

a second degradable polyolefin layer disposed on said first degradable tie layer;

a third degradable polyolefin layer disposed on said second degradable polyolefin layer;

a second degradable tie layer disposed on said third degradable polyolefin layer;

a moisture protectant layer disposed on said inside surface of said substrate including a biodegradable polyester selected from the group consisting of polyvinyl alcohol, a polyhydroxyalkanoate, and polylactic acid;

a third degradable tie layer disposed on said moisture protectant layer; and

a fourth degradable polyolefin layer disposed on said third degradable tie layer.

41. A laminate for forming moisture barriers, comprising:

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a substrate having an interior surface for contacting products; and

a moisture protectant layer disposed on said interior surface of said substrate and including a degradable polymer selected from the group consisting of a degradable polyolefin, a polyhydroxyalkanoate, and polylactic acid, and polyester.

42. The laminate according to claim 41, wherein said moisture protectant layer has a coating thickness from five to thirty pounds per three-thousand square feet.

43. A drinking cup for hot liquids, comprising the laminate according to claim 41.

44. A laminate for forming moisture barriers, comprising:

a substrate having an interior surface for contacting products and an exterior surface; and

a first moisture protectant layer disposed on said interior surface of said substrate and including a degradable polymer selected from the group consisting of a degradable polyolefin, a polyhydroxyalkanoate, and polylactic acid, and polyester; and

a second moisture protectant layer disposed on said exterior surface of said substrate and including a degradable polymer selected from the group consisting of a degradable polyolefin, a polyhydroxyalkanoate, polylactic acid, and polyester.

45. The laminate according to claim 44, wherein said first and second moisture protectant layers each have a coating thickness from five to thirty pounds per three-thousand square feet.

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46. A packaging for cold, moisture-sensitive contents, comprising the laminate according to claim 44.

47. A gable top carton for storing a non-oxygen sensitive product, comprising the laminate according to claim 44.

48. The gable top carton according to claim 37, wherein the non-oxygen sensitive product is pasteurized milk.

49. A pouch top carton for storing non-oxygen sensitive products, comprising the laminate according to claim 44.

50. The laminate according to claim 1, wherein at least one of said layers includes calcium carbonate in order to increase a degradation rate.

51. The laminate according to claim 1, wherein at least one of said layers includes a filler for increasing a degradation rate, said filler being selected from the group consisting of starch, clay, kaolin, talc, cellulose fiber, and diatomaceous earth.

52. The laminate according to claim 1, wherein at least one of said layers include an antioxidant for removing low molecular-weight products.

53. The laminate according to claim 52, wherein said antioxidant is selected from the group consisting of vitamin E and tetrakis-(methylene-(3,5-di-terbutyl-4-hydrocinnamate)methane.

54. The laminate according to claim 52, wherein said antioxidant forms at most five weight percent of any given one of said layers.

55. The laminate according to claim 1, wherein at least one of said layers includes a zeolite for removing low molecular-weight products.

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56. The laminate according to claim 55, wherein said zeolite forms up to five weight percent of any given one of said layers.

57. The laminate according to claim 1, including said degradable tie layer, said degradable tie layer including a metal catalyst and a tie resin.

58. The laminate according to claim 57, wherein said tie resin is ethylene-co-maleic anhydride.

59. A method for making biodegradable paper products with barrier properties, which comprises:

providing a substrate having a surface; and

extruding a barrier layer onto said surface of said substrate, said barrier layer including a degradable material selected from the group consisting of polylactic acid and a polyhydroxyalkonoate.

60. The method according to claim 59, which further comprises controlling a coatweight of said degradable material within a range from 8 to 20 pounds per three-thousand square feet.

61. The method according to claim 59, which further comprises extruding a second barrier layer on an opposing side of said substrate, said second barrier layer including a degradable material selected from the group consisting of polylactic acid and a polyhydroxyalkonoate.

62. The method according to claim 59, which further comprises applying primer to said substrate before the extruding step.

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63. The method according to claim 62, which further comprises:

using an aqueous primer as the primer; and

removing water from the aqueous primer immediately prior the extruding step.

64. The method according to claim 62, which further comprises selecting said primer from a group consisting of a starch emulsion, a soy protein emulsion, polyvinyl acetate, and polyvinyl alcohol.

65. The method according to claim 62, which further comprises controlling a coatweight of said primer in a range from 0.5 to 5 pounds per three-thousand square feet.

66. The method according to claim 61, which further comprises adding a filler to said degradable material before the extruding step.

67. The method according to claim 66, which further comprises selecting said filler from the group consisting of calcium carbonate, talc, diatomaceous earth, and clay.

68. The method according to claim 66, which further comprises controlling an amount of said filler in said degradable material in a range from 5 to 20 percent by weight.

69. The method according to claim 59, which further comprises, when polyhydroxyalkanoate is said degradable material, adding an antiblock material to said barrier layer after the extruding step.

70. The method according to claim 69, which further comprises selecting said antiblock material from the group consisting of erucamide, calcium carbonate, and talc.

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71. The method according to claim 69, which further comprises controlling an amount of said antiblock material in a range from 0.1 to 5 percent by weight.

72. A folding carton comprising the laminate according to claim 44.

73. A corrugated box comprising the laminate according to claim 44.

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Abstract of the Disclosure:

Biodegradable laminates including biodegradable polymers have oxygen and moisture barrier properties. The oxygen and moisture barrier layers are biodegradable. If the barrier layers have poor adhesion, degradable tie layers can be included to improve adhesion. Combination of laminate structures can be formed to increase the overall oxygen and moisture barrier characteristics. The biodegradable laminates can be formed by extrusion.

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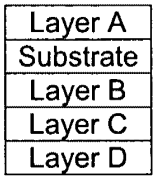

Layer A
Substrate
Layer B
Layer C
Layer D

Fig. 1

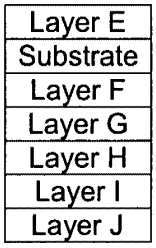

Layer E
Substrate
Layer F
Layer G
Layer H
Layer I
Layer J

Fig. 2

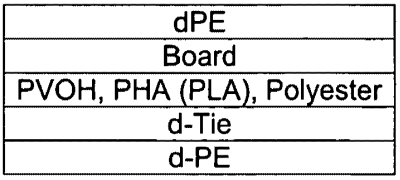


Fig. 3A

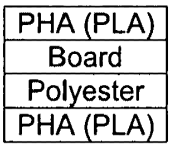


Fig. 3B

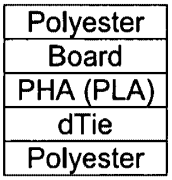


Fig. 3C

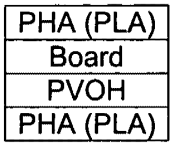


Fig. 4A

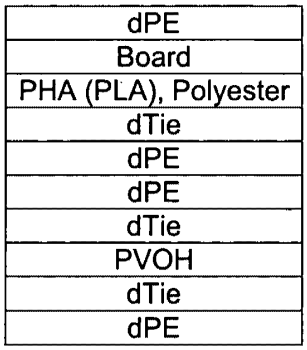


Fig. 4B

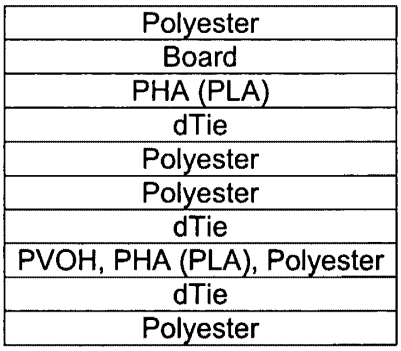


Fig. 4C

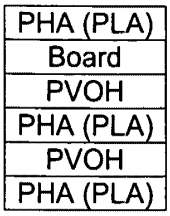


Fig. 5A

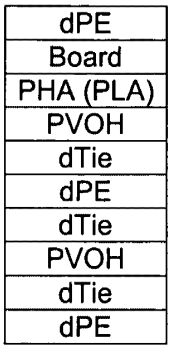


Fig. 5B

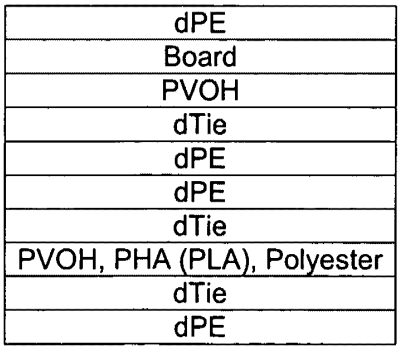


Fig. 5C

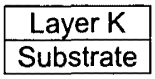


Fig. 6

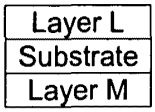


Fig. 7

PATENT APPLICATION SERIAL NO. _____

U.S. DEPARTMENT OF COMMERCE
PATENT AND TRADEMARK OFFICE
FEE RECORD SHEET

02/08/2006 AKELECH1 00000046 090525 11348150

01 FC:1011 300.00 DA
02 FC:1111 500.00 DA
03 FC:1311 200.00 DA
04 FC:1201 2000.00 DA
05 FC:1202 2650.00 DA

PTO-1556
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*U.S. Government Printing Office: 2002 — 466-267/68033

PTO/SB/06 (12-04)
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PATENT APPLICATION FEE DETERMINATION RECORD

Substitute for Form PTO-875 Effective December 8, 2004

Application or Docket Number: 11348150

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	150.00		N/A	300.00
SEARCH FEE (37 CFR 1.16(k), (l), or (m))	N/A	N/A	N/A	$250		N/A	$500
EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A	N/A	$100		N/A	$200
TOTAL CLAIMS (37 CFR 1.16(i))	73 minus 20 =	* 53	X$ 25 =		OR	X$50 =	2650.
INDEPENDENT CLAIMS (37 CFR 1.16(h))	13 minus 3 =	* 10	X100 =			X200 =	2000.
APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $250 ($125 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))			+180=			+360=	
* If the difference in column 1 is less than zero, enter "0" in column 2.			TOTAL			TOTAL	5650

1, 32, 33, 34, 35, 36, 37, 18, 39, 40, 41, 44, 59

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$ 25 =		OR	X$50 =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=	X100 =		OR	X200 =	
	Application Size Fee (37 CFR 1.16(s))									
	FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))					+180=		OR	+360=	
						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	RATE ($)	ADDITIONAL FEE ($)	OR	RATE ($)	ADDITIONAL FEE ($)
AMENDMENT B	Total (37 CFR 1.16(i))	*	Minus	**	=	X$ 25 =		OR	X$50 =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=	X100 =		OR	X200 =	
	Application Size Fee (37 CFR 1.16(s))									
	FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))					+180=		OR	+360=	
						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 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.

Application Data Sheet

Application Information

Application Type::	Regular
Subject Matter::	Utility
Title::	BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOISTURE BARRIER PROPERTIES AND METHOD FOR MAKING BIODEGRADABLE PAPER-BASED LAMINATE
Attorney Docket Number::	FDS-029388-US
Request for Early Publication?::	No
Request for Non-Publication?::	No
Total Drawing Sheets::	5
Small Entity::	No

Applicant Information

Applicant Authority Type::	Inventor
Primary Citizenship Country::	US
Status::	Full Capacity
Given Name::	CHRISTOPHER
Middle Name::	S.
Family Name::	CLEVELAND
City of Residence::	NEWPORT
State/Province of Residence::	KY
Country of Residence::	US
Street of Mailing Address::	828 MAPLE AVENUE
City of Mailing Address::	NEWPORT
State/Province of Mailing Address::	KY
Country of Mailing Address::	US
Postal/Zip Code of Mailing Address::	41071

Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
Page 39 of 1179

Applicant Information

Applicant Authority Type::	Inventor
Primary Citizenship Country::	US
Status::	Full Capacity
Given Name::	TRICIA
Middle Name::	S.
Family Name::	REIGHARD
City of Residence::	LOVELAND
State/Province of Residence::	OH
Country of Residence::	US
Street of Mailing Address::	9874 STONEBRIDGE DRIVE
City of Mailing Address::	LOVELAND
State/Province of Mailing Address::	OH
Country of Mailing Address::	US
Postal/Zip Code of Mailing Address::	45140

Applicant Information

Applicant Authority Type::	Inventor
Primary Citizenship Country::	US
Status::	Full Capacity
Given Name::	JAMES
Middle Name::	I.
Family Name::	MARCHMAN
City of Residence::	LOVELAND
State/Province of Residence::	OH
Country of Residence::	US
Street of Mailing Address::	4045 CEDARWOOD LANE
City of Mailing Address::	LOVELAND
State/Province of Mailing Address::	OH
Country of Mailing Address::	US
Postal/Zip Code of Mailing Address::	45140

Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
Page 40 of 1179

Correspondence Information

Correspondence Customer Number::	01726
Phone Number::	513/248-6069
Fax Number::	513/248-6455
E-Mail Address::	melvin.fletcher@ipaper.com

Representative Information

Representative Customer Number::	01726

Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
Page 41 of 1179

18304 U.S. PTO
020606

PTO/SB/05 (04-05)
Approved for use through 07/31/2006. 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.

UTILITY PATENT APPLICATION TRANSMITTAL

(Only for new nonprovisional applications under 37 CFR 1.53(b))

Attorney Docket No.	FDS-029388-US
First Inventor	CLEVELAND, et al.
Title	BIODEGARADABLE PAPER-BASED ...
Express Mail Label No.	EQ239461176US

112587 U.S. PTO
020606

APPLICATION ELEMENTS

See MPEP chapter 600 concerning utility patent application contents.

1. ☑ **Fee Transmittal Form** (e.g., PTO/SB/17)
   (Submit an original and a duplicate for fee processing)
2. ☐ **Applicant claims small entity status.**
   See 37 CFR 1.27.
3. ☑ **Specification** [*Total Pages* 28]
   Both the claims and abstract must start on a new page
   (For information on the preferred arrangement, see MPEP 608.01(a))
4. ☑ **Drawing(s)** (*35 U.S.C. 113*) [*Total Sheets* 5]
5. **Oath or Declaration** [*Total Sheets* _____]
   a. ☐ Newly executed (original or copy)
   b. ☐ A copy from a prior application (37 CFR 1.63(d))
   (for continuation/divisional with Box 18 completed)
   i. ☐ DELETION OF INVENTOR(S)
   Signed statement attached deleting inventor(s) name in the prior application, see 37 CFR 1.63(d)(2) and 1.33(b).
6. ☑ **Application Data Sheet.** See 37 CFR 1.76
7. ☐ **CD-ROM or CD-R** in duplicate, large table or Computer Program *(Appendix)*
   ☐ Landscape Table on CD
8. **Nucleotide and/or Amino Acid Sequence Submission**
   (if applicable, items a. – c. are required)
   a. ☐ Computer Readable Form (CRF)
   b. Specification Sequence Listing on:
      i. ☐ CD-ROM or CD-R (2 copies); or
      ii. ☐ Paper
   c. ☐ Statements verifying identity of above copies

ADDRESS TO:
Commissioner for Patents
P.O. Box 1450
Alexandria VA 22313-1450

ACCOMPANYING APPLICATION PARTS

9. ☐ **Assignment Papers** (cover sheet & document(s))
   Name of Assignee _____
10. ☐ **37 CFR 3.73(b) Statement** *(when there is an assignee)* ☐ **Power of Attorney**
11. ☐ **English Translation Document** *(if applicable)*
12. ☐ **Information Disclosure Statement** (PTO/SB/08 or PTO-1449)
    ☐ Copies of citations attached
13. ☐ **Preliminary Amendment**
14. ☑ **Return Receipt Postcard** (MPEP 503)
    (Should be specifically itemized)
15. ☐ **Certified Copy of Priority Document(s)**
    (if foreign priority is claimed)
16. ☐ **Nonpublication Request** under 35 U.S.C. 122(b)(2)(B)(i).
    Applicant must attach form PTO/SB/35 or equivalent.
17. ☑ Other: cover letter

18. If a CONTINUING APPLICATION, *check appropriate box, and supply the requisite information below and in the first sentence of the specification following the title, or in an Application Data Sheet under 37 CFR 1.76:*

☐ Continuation ☐ Divisional ☐ Continuation-in-part (CIP) of prior application No.: .....

Prior application information: *Examiner* _____ *Art Unit:* _____

19. CORRESPONDENCE ADDRESS

☑ The address associated with Customer Number: 01726 *OR* ☐ Correspondence address below

Name					
Address					
City		State		Zip Code	
Country		Telephone		Email	

Signature	Melvin D. Fletcher	Date	FEBRUARY 6, 2006
Name (Print/Type)	MELVIN D. FLETCHER	Registration No. (Attorney/Agent)	36,137

This collection of information is required by 37 CFR 1.53(b). 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. 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.

020606 18304 U.S. PTO

PTO/SB/17 (12-04v2)
Approved for use through 07/31/2006. 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.

Effective on 12/08/2004.
Fees pursuant to the Consolidated Appropriations Act, 2005 (H.R. 4818).

FEE TRANSMITTAL
For FY 2005

☐ Applicant claims small entity status. See 37 CFR 1.27

TOTAL AMOUNT OF PAYMENT	($)	5650.00

Complete if Known	
Application Number	NEW APPLICATION
Filing Date	FEBRUARY 6, 2006
First Named Inventor	CLEVELAND, et al.
Examiner Name	
Art Unit	
Attorney Docket No.	FDS-029388-US

METHOD OF PAYMENT (check all that apply)

☐ Check ☐ Credit Card ☐ Money Order ☐ None ☐ Other (please identify): _____

☑ Deposit Account Deposit Account Number: 09-0525 Deposit Account Name: INTL PAPER CO

For the above-identified deposit account, the Director is hereby authorized to: (check all that apply)

☑ Charge fee(s) indicated below ☐ Charge fee(s) indicated below, except for the filing fee

☑ Charge any additional fee(s) or underpayments of fee(s) under 37 CFR 1.16 and 1.17 ☑ Credit any overpayments

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.

FEE CALCULATION

1. BASIC FILING, SEARCH, AND EXAMINATION FEES

	FILING FEES		SEARCH FEES		EXAMINATION FEES		
Application Type	Fee ($)	Small Entity Fee ($)	Fee ($)	Small Entity Fee ($)	Fee ($)	Small Entity Fee ($)	Fees Paid ($)
Utility	300	150	500	250	200	100	1000.00
Design	200	100	100	50	130	65	
Plant	200	100	300	150	160	80	
Reissue	300	150	500	250	600	300	
Provisional	200	100	0	0	0	0	

2. EXCESS CLAIM FEES

Fee Description	Fee ($)	Small Entity Fee ($)
Each claim over 20 (including Reissues)	50	25
Each independent claim over 3 (including Reissues)	200	100
Multiple dependent claims	360	180

Total Claims	Extra Claims	Fee ($)	Fee Paid ($)
73 - 20 or HP =	53	x 50.00	= 2650.00

HP = highest number of total claims paid for, if greater than 20.

Indep. Claims	Extra Claims	Fee ($)	Fee Paid ($)
13 - 3 or HP =	10	x 200.00	= 2000.00

HP = highest number of independent claims paid for, if greater than 3.

Multiple Dependent Claims	
Fee ($)	Fee Paid ($)

3. APPLICATION SIZE FEE

If the specification and drawings exceed 100 sheets of paper (excluding electronically filed sequence or computer listings under 37 CFR 1.52(e)), the application size fee due is $250 ($125 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).

Total Sheets	Extra Sheets	Number of each additional 50 or fraction thereof	Fee ($)	Fee Paid ($)
_____ - 100 =	_____ / 50 =	_____ (round up to a whole number)	x _____	= _____

4. OTHER FEE(S)

	Fees Paid ($)
Non-English Specification, $130 fee (no small entity discount)	
Other (e.g., late filing surcharge): _____	

SUBMITTED BY

Signature	Melvin D. Fletcher	Registration No. (Attorney/Agent) 36,137	Telephone 513/248-6069
Name (Print/Type)	MELVIN D. FLETCHER		Date 2/6/2006

This collection of information is required by 37 CFR 1.136. 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, 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.

INTERNATIONAL PAPER

MELVIN D. FLETCHER
SENIOR COUNSEL
INTELLECTUAL PROPERTY

CINCINNATI TECHNOLOGY CENTER
6285 TRI-RIDGE BOULEVARD
LOVELAND OH 45140

T 513 248 6069
F 513 248 6455
melvin.fletcher@ipaper.com

VIA USPS EXPRESS MAIL
LABEL NO. EQ239461176US

February 6, 2006

Commissioner for Patents
Post Office Box 1450
Alexandria, Virginia 22313-1450

RE: SUBMISSION OF NON-PROVISIONAL APPLICATION
Applicant : CLEVELAND, et al.
Title : BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN.....
Our Ref. : FDS-029388-US

Dear Commissioner:

Enclosed herewith for filing, Applicant(s) respectfully submit(s) the following checked items:

- [x] Utility Patent Application Transmittal, Form PTO/SB/05, 1 sheet(s);
- [x] Fee Transmittal, Form PTO/SB/17, 1 sheet(s);
- [x] Application Data Sheet, 3 sheet(s);
- [x] Patent Specification, 28 sheet(s);
- [x] 5 Figures arranged on 5 sheet(s);
- [ ] Information Disclosure Statement, Form PTO/SB/08, _____ sheet(s), and one copy of each reference cited thereon;
- [ ] Signed Inventors Declaration(s), _____ sheet(s);
- [ ] Nonpublication Request, Form PTO/SB/35, _____ sheet(s); and,
- [x] a postage-prepaid, return-receipt postcard with a listing of the above-checked items.

Please stamp the enclosed postcard and return same to me to indicate your receipt of the above-listed items. Please feel free to contact me if you have any questions concerning the above or the enclosed.

With kindest regards,

Melvin D. Fletcher
Attorney for Applicant(s)

Enclosures
MDF/ jat

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	ATTORNEY DOCKET NUMBER
11/348,150	02/06/2006	Christopher S. Cleveland	FDS-029388-US

CONFIRMATION NO. 5681

01726
INTERNATIONAL PAPER COMPANY
6285 TRI-RIDGE BOULEVARD
LOVELAND, OH 45140

FORMALITIES LETTER

Date Mailed: 03/07/2006

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 and pay any fees required 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 oath or declaration is missing. *A properly signed oath or declaration in compliance with 37 CFR 1.63, identifying the application by the above Application Number and Filing Date, is required.*
  Note: If a petition under 37 CFR 1.47 is being filed, an oath or declaration in compliance with 37 CFR 1.63 signed by all available joint inventors, or if no inventor is available by a party with sufficient proprietary interest, is required.

The applicant needs to satisfy supplemental fees problems indicated below.

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

• To avoid abandonment, a surcharge (for late submission of filing fee, search fee, examination fee or oath or declaration) as set forth in 37 CFR 1.16(f) of $130 for a non-small entity, must be submitted with the missing items identified in this letter.

SUMMARY OF FEES DUE:

Total additional fee(s) required for this application is **$130** for a Large Entity

- **$130** Surcharge.

Replies should be mailed to: Mail Stop Missing Parts
Commissioner for Patents

P.O. Box 1450
Alexandria VA 22313-1450

A copy of this notice MUST be returned with the reply.

Office of Initial Patent Examination (571) 272-4000, or 1-800-PTO-9199, or 1-800-972-6382
PART 3 - OFFICE COPY

RECEIVED
CENTRAL FAX CENTER
MAR 15 2006

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	ATTORNEY DOCKET NUMBER
11/348,150	02/06/2006	Christopher S. Cleveland	FDS-029388-US

CONFIRMATION NO. 5681

01726
INTERNATIONAL PAPER COMPANY
6285 TRI-RIDGE BOULEVARD
LOVELAND, OH 45140

FORMALITIES LETTER

Date Mailed: 03/07/2006

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 and pay any fees required 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 oath or declaration is missing. *A properly signed oath or declaration in compliance with 37 CFR 1.63, identifying the application by the above Application Number and Filing Date, is required.*
  Note: If a petition under 37 CFR 1.47 is being filed, an oath or declaration in compliance with 37 CFR 1.63 signed by all available joint inventors, or if no inventor is available by a party with sufficient proprietary interest, is required.

The applicant needs to satisfy supplemental fees problems indicated below.

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

- To avoid abandonment, a surcharge (for late submission of filing fee, search fee, examination fee or oath or declaration) as set forth in 37 CFR 1.16(f) of $130 for a non-small entity, must be submitted with the missing items identified in this letter.

SUMMARY OF FEES DUE:

Total additional fee(s) required for this application is **$130** for a Large Entity

- **$130** Surcharge.

Replies should be mailed to: Mail Stop Missing Parts
Commissioner for Patents

03/17/2006 EFLORES 00000086 090525 11348150
01 FC:1051 130.00 DA

P.O. Box 1450
Alexandria VA 22313-1450

A copy of this notice MUST be returned with the reply.

Office of Initial Patent Examination (571) 272-4000, or 1-800-PTO-9199, or 1-800-972-6382
PART 2 - COPY TO BE RETURNED WITH RESPONSE

PAGE 5/12 * RCVD AT 3/15/2006 1:35:49 PM [Eastern Standard Time] * SVR:USPTO-EFXRF-2/8 * DNIS:2738300 * CSID:5132486680 * DURATION (mm-ss):05-18

PTO/SB/17 (12-04v2)
Approved for use through 07/31/2006. 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.

RECEIVED
CENTRAL FAX CENTER
MAR 15 2006

Effective on 12/08/2004.
Fees pursuant to the Consolidated Appropriations Act, 2005 (H.R. 4818).

FEE TRANSMITTAL
For FY 2005

☐ Applicant claims small entity status. See 37 CFR 1.27

TOTAL AMOUNT OF PAYMENT	($)	130.00

Complete if Known	
Application Number	11/348,150
Filing Date	FEBRUARY 6, 2006
First Named Inventor	CLEVELAND et al.
Examiner Name	
Art Unit	1772
Attorney Docket No.	FDS-029388-US

METHOD OF PAYMENT (check all that apply)

☐ Check ☐ Credit Card ☐ Money Order ☐ None ☐ Other (please identify): _____

☑ Deposit Account Deposit Account Number: 09-0525 Deposit Account Name: INTL PAPER CO

For the above-identified deposit account, the Director is hereby authorized to: (check all that apply)

☑ Charge fee(s) indicated below ☐ Charge fee(s) indicated below, except for the filing fee

☑ Charge any additional fee(s) or underpayments of fee(s) under 37 CFR 1.16 and 1.17 ☑ Credit any overpayments

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.

FEE CALCULATION

1. BASIC FILING, SEARCH, AND EXAMINATION FEES

Application Type	FILING FEES Fee ($)	FILING FEES Small Entity Fee ($)	SEARCH FEES Fee ($)	SEARCH FEES Small Entity Fee ($)	EXAMINATION FEES Fee ($)	EXAMINATION FEES Small Entity Fee ($)	Fees Paid ($)
Utility	300	150	500	250	200	100	
Design	200	100	100	50	130	65	
Plant	200	100	300	150	160	80	
Reissue	300	150	500	250	600	300	
Provisional	200	100	0	0	0	0	

2. EXCESS CLAIM FEES

Fee Description	Fee ($)	Small Entity Fee ($)
Each claim over 20 (including Reissues)	50	25
Each independent claim over 3 (including Reissues)	200	100
Multiple dependent claims	360	180

Total Claims _____ - 20 or HP = _____ x Extra Claims _____ Fee ($) _____ = Fee Paid ($) _____

HP = highest number of total claims paid for, if greater than 20.

Indep. Claims _____ - 3 or HP = _____ x Extra Claims _____ Fee ($) _____ = Fee Paid ($) _____

HP = highest number of independent claims paid for, if greater than 3.

Multiple Dependent Claims: Fee ($) _____ Fee Paid ($) _____

3. APPLICATION SIZE FEE

If the specification and drawings exceed 100 sheets of paper (excluding electronically filed sequence or computer listings under 37 CFR 1.52(e)), the application size fee due is $250 ($125 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).

Total Sheets _____ - 100 = _____ Extra Sheets _____ / 50 = _____ Number of each additional 50 or fraction thereof (round up to a whole number) x Fee ($) _____ = Fee Paid ($) _____

4. OTHER FEE(S)

	Fees Paid ($)
Non-English Specification, $130 fee (no small entity discount)	
Other (e.g., late filing surcharge): SURCHARGE	130.00

SUBMITTED BY

Signature	Melvin D. Fletcher	Registration No. (Attorney/Agent) 36,137	Telephone 513-248-6069
Name (Print/Type)	MELVIN D. FLETCHER		Date 3/15/2006

This collection of information is required by 37 CFR 1.138. 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, 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.

INTERNATIONAL PAPER

CINCINNATI TECHNOLOGY CENTER
6285 TRI-RIDGE BOULEVARD
LOVELAND OH 45140

T 513 248 6000
F 513 248 6455

RECEIVED
CENTRAL FAX CENTER
MAR 15 2006

CONFIDENTIAL FACSIMILE TRANSMISSION

To: Commissioner for Patents
Fax: 571-273-8300
Tel:
Date: March 15, 2006
RE: Missing Parts

Our File Ref.: FDS-029388-US
Your File Ref.: 11/348,150
No. of Pages: 12 (including this cover sheet)

☐ URGENT ☐ FOR YOUR INFORMATION

☐ CONFIRMATION COPY FOLLOWS BY:
☐ FIRST-CLASS U.S. MAIL
☐ OVERNIGHT MAIL
☐ INTERNATIONAL MAIL

☐ NO COPY FOLLOWS

In response to the Office Action dated 3-7-2006, please find the following checked items:

☑ Response to Notice to File Missing Parts of Nonprovisional Application, 2 sheet(s);

☐ Petition for Extension of Time Under 37 C.F.R. 1.136(a), Form PTO/SB/22, and one (1) copy thereof attached thereto, _____ sheet(s);

☑ Copy of Notice to File Missing Parts of Nonprovisional Application, 2 sheet(s);

☑ Fee Transmittal, Form PTO/SB/17, 1 sheet(s);

☑ Declaration(s) Under 37 C.F.R. 1.63 and Appointment of Power of Attorney signed by Applicant(s), 6 sheet(s);

☐ Information Disclosure Statement by Applicant(s), _____ sheet(s), to which Form PTO/SB/08, _____ sheet(s), has been attached along with a copy of each reference(s) cited thereon.

☐ Other:

Thank you.

Certificate of Transmission Under 37 C.F.R. 1.8

The undersigned hereby certifies that a true and accurate copy of the items checked above are being transmitted to the Honorable Commissioner for Patents, by facsimile transmission to the facsimile number indicated above, on this the 15TH day of MARCH, 20 06.

JANE A. TOMLINSON

This facsimile transmission may contain confidential and/or legally privileged information from the law department of International Paper Company which is intended only for the use of the individual(s) named above. If you are not the intended recipient, you are hereby notified that any disclosure, copying or distribution of this facsimile transmission is strictly prohibited. If you have received this transmission in error, please notify us immediately by phoning (513) 248-6207 so that we can arrange for the return of the documents. Thank you.

INTERNATIONAL PAPER

MELVIN D. FLETCHER
SENIOR COUNSEL
INTELLECTUAL PROPERTY

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LOVELAND OH 45140

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SENT ☒ **BY FACSIMILE TO (571-273-8300)**
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March 15, 2006

Commissioner for Patents
United States Patent and Trademark Office
Post Office Box 1450
Alexandria, Virginia 22313-1450

UNITED STATES PATENT AND TRADEMARK OFFICE
WASHINGTON, D.C., UNITED STATES OF AMERICA

In re Application of:

Applicant(s): CLEVELAND et al.

Serial No.: 11/348,150

Filed on: FEBRUARY 6, 2006

Entitled: BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOISTURE BARRIER PROPERTIES AND METHOD FOR MAKING BIODEGRADABLE .....

Examiner:

Our File Ref.: FDS-029388-US

Group Art Unit: 1772

RESPONSE TO NOTICE TO FILE MISSING PARTS OF NONPROVISIONAL APPLICATION - FILING DATE GRANTED

Now comes Applicant, by counsel, and in response to the Notice dated 03/07/2006, respectfully submits the following checked items:

☐ Petition for Extension of Time Under 37 C.F.R. 1.136(a), Form PTO/SB/22, and one (1) copy thereof attached thereto, _____ sheet(s);

☒ Copy of Notice to File Missing Parts of Nonprovisional Application, 2 sheet(s);

☒ Fee Transmittal, Form PTO/SB/17, 1 sheet(s);

INTERNATIONAL PAPER

- [x] Declaration(s) Under 37 C.F.R. 1.63 and Appointment of Power of Attorney signed by Applicant(s), 6 sheet(s);
- [ ] Information Disclosure Statement by Applicant(s), _____ sheet(s), to which Form PTO/SB/08, _____ sheet(s), has been attached along with a copy of each reference(s) cited thereon;
- [ ] Other:
- [ ] Postage-prepaid return-receipt postcard for your use in stamping to indicate receipt of the above-listed items.

Should the Office have any questions concerning the above or the enclosed, Applicant(s) respectfully request(s) that the Office contact the undersigned attorney, who can be reached at the telephone numbers and email address listed.

Respectfully submitted,

Date: March 15, 2006

Melvin D. Fletcher, Reg. No. 36,167
Counsel for Applicant(s)

International Paper Company
6285 Tri-Ridge Boulevard
Loveland, Ohio 45140
513.248.6069 phone
513.248.6455 fax

Certification of Mailing or Transmission Under 37 C.F.R. 1.8

The undersigned hereby certifies that a true and accurate copy of the within "Response to Notice to File Missing Parts of Nonprovisional Application", together with all attachments referred-to herein, is being transmitted to the Honorable Commissioner for Patents, either by first-class mail, postage prepaid, addressed to Commissioner for Patents, Post Office Box 1450, Alexandria, Virginia 22313-1450, or by facsimile transmission to the facsimile number indicated hereon, on this the 15th day of March, 20 06.

JANE A. TOMLINSON

DECLARATION UNDER 37 C.F.R. 1.63 AND APPOINTMENT OF POWER OF ATTORNEY

As a below-named inventor, I hereby declare the following:

- My residence, post office address and citizenship are as stated below, next to my name.
- I believe that I am an original, first and sole inventor (if only one name is listed below) or joint inventor (if more than one name is listed below) of the subject matter which is claimed and for which a patent is sought on an invention entitled, " BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOISTURE BARRIER PROPERTIES AND METHOD..... ", the specification of which:

  ☐ is attached hereto
  ☒ was filed on 2/6/2006
  as application no. 11/348,150
  and was amended on _____

- I authorize my below-named attorneys/agents to enter in the spaces above, either before or after I have executed this declaration, whether the invention is described and claimed in an attached specification and to provide and enter the filing date and serial number of any corresponding US patent application, if previously filed, as well as the date of any amendment made thereto, without further authorization from me.
- 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 above.
- I hereby acknowledge my duty to disclose information which may be material to patentability, as defined in Title 37 of the Code of Federal Regulations, Section 1.56.
- I hereby claim foreign priority benefits under Title 35, United States Code, Sections 119(a)-(d), of any foreign application(s) for patent or inventor's certificate(s) 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 upon which priority is claimed:

Foreign Priority Application(s)			Priority Claimed?	
(Serial Number)	(Country)	(Filing Date - Month/Day/Year)	(Yes)	(No)
(Serial Number)	(Country)	(Filing Date - Month/Day/Year)	(Yes)	(No)
(Serial Number)	(Country)	(Filing Date - Month/Day/Year)	(Yes)	(No)

- I hereby claim the benefit under Title 35, United States Code, Section 120, of any prior U.S. patent application(s) listed below and, insofar as the subject matter of each of the claims of the above-referenced specification is not disclosed in the prior U.S. patent application(s) in a manner provided by the first paragraph of Title 35, United States Code, Section 112, I acknowledge the duty to disclose material information, as defined in Title 37, Code of Federal Regulations, Section 1.56, which occurred between the filing date of the prior application(s) and the national or PCT international filing date of the application for patent made upon the above-referenced specification:

Prior Application(s)		Status
(Serial Number)	(Filing Date - Month/Day/Year)	(pending, granted, abandoned)
(Serial Number)	(Filing Date - Month/Day/Year)	(pending, granted, abandoned)
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Attorney Docket No. FDS-029388-US

PAGE 7/12 * RCVD AT 3/15/2006 1:35:49 PM [Eastern Standard Time] * SVR:USPTO-EFXRF-2/8 * DNIS:2738300 * CSID:5132486680 * DURATION (mm-ss):05-18

DECLARATION UNDER 37 C.F.R. 1.63 AND APPOINTMENT OF POWER OF ATTORNEY

- I hereby claim domestic priority benefits under Title 35, United States Code, Section 119(e), of any provisional patent application(s) listed below:

Provisional Application(s)	Filing Date
(Serial Number)	(Month/Day/Year)
(Serial Number)	(Month/Day/Year)
(Serial Number)	(Month/Day/Year)

- 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 knowledge that willful false statements and the like are made punishable by fine or imprisonment, or both, under Title 18, United States Code, Section 1001, and that such willful false statements may jeopardize the validity of any application for patent made upon the above-referenced specification, or of any patent issuing therefrom.
- I hereby appoint the individuals associated with the following Customer Number to act as my attorneys/agents, to prosecute and to transact all business in the United States Patent and Trademark Office connected therewith:

USPTO Customer Number 01726

- I hereby request that all communications concerning any application for patent made upon the above-referenced specification be sent to the correspondence address associated with the above-stated Customer Number.

Signature of First Inventor — Date: 2-23-06

CHRISTOPHER S. CLEVELAND — Typed Name of First Inventor — Country of Citizenship: US

828 MAPLE AVENUE, NEWPORT, KY. 41071 — Residence and/or Mailing Address (the same if only one address is listed, please indicate separately if not the same)

Signature of Second Inventor — Date

TRICIA S. REIGHARD — Typed Name of Second Inventor — Country of Citizenship: US

9874 STONEBRIDGE DRIVE, LOVELAND, OH. 45140 — Residence and/or Mailing Address (the same if only one address is listed, please indicate separately if not the same)

Signature of Third Inventor — Date

JAMES I. MARCHMAN — Typed Name of Third Inventor — Country of Citizenship: US

4045 CEDARWOOD LANE, LOVELAND, OH. 45140 — Residence and/or Mailing Address (the same if only one address is listed, please indicate separately if not the same)

PAGE 8/12 * RCVD AT 3/15/2006 1:35:49 PM [Eastern Standard Time] * SVR:USPTO-EFXRF-2/8 * DNIS:2738300 * CSID:5132486680 * DURATION (mm-ss):05-18

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  ☐ is attached hereto
  ☒ was filed on 2/6/2006
  as application no. 11/348,150
  and was amended on _____

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(Serial Number)	(Country)	(Filing Date - Month/Day/Year)	(Yes)	(No)
(Serial Number)	(Country)	(Filing Date - Month/Day/Year)	(Yes)	(No)
(Serial Number)	(Country)	(Filing Date - Month/Day/Year)	(Yes)	(No)

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Provisional Application(s)	Filing Date
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(Serial Number)	(Month/Day/Year)

- 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 knowledge that willful false statements and the like are made punishable by fine or imprisonment, or both, under Title 18, United States Code, Section 1001, and that such willful false statements may jeopardize the validity of any application for patent made upon the above-referenced specification, or of any patent issuing therefrom.
- I hereby appoint the individuals associated with the following Customer Number to act as my attorneys/agents, to prosecute and to transact all business in the United States Patent and Trademark Office connected therewith:

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Signature of First Inventor — Date

CHRISTOPHER S. CLEVELAND — US
Typed Name of First Inventor — Country of Citizenship

828 MAPLE AVENUE, NEWPORT, KY. 41071
Residence and/or Mailing Address (the same if only one address is listed, please indicate separately if not the same)

Tricia S. Reighard — 2/27/2006
Signature of Second Inventor — Date

TRICIA S. REIGHARD — US
Typed Name of Second Inventor — Country of Citizenship

9874 STONEBRIDGE DRIVE, LOVELAND, OH. 45140
Residence and/or Mailing Address (the same if only one address is listed, please indicate separately if not the same)

Signature of Third Inventor — Date

JAMES I. MARCHMAN — US
Typed Name of Third Inventor — Country of Citizenship

4045 CEDARWOOD LANE, LOVELAND, OH. 45140
Residence and/or Mailing Address (the same if only one address is listed, please indicate separately if not the same)

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  ☒ was filed on 2/6/2006
  as application no. 11/348,150
  and was amended on _____

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(Serial Number)	(Filing Date - Month/Day/Year)	(pending, granted, abandoned)
(Serial Number)	(Filing Date - Month/Day/Year)	(pending, granted, abandoned)

Attorney Docket No. FDS-029388-US

PAGE 11/12 * RCVD AT 3/15/2006 1:35:49 PM [Eastern Standard Time] * SVR:USPTO-EFXRF-2/8 * DNIS:2738300 * CSID:5132486680 * DURATION (mm-ss):05-18

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CHRISTOPHER S. CLEVELAND — US

Typed Name of First Inventor — Country of Citizenship

828 MAPLE AVENUE, NEWPORT, KY. 41071

Residence and/or Mailing Address (the same if only one address is listed, please indicate separately if not the same)

Signature of Second Inventor — Date

TRICIA S. REIGHARD — US

Typed Name of Second Inventor — Country of Citizenship

9874 STONEBRIDGE DRIVE, LOVELAND, OH. 45140

Residence and/or Mailing Address (the same if only one address is listed, please indicate separately if not the same)

James I. Marchman — 3/9/2006

Signature of Third Inventor — Date

JAMES I. MARCHMAN — US

Typed Name of Third Inventor — Country of Citizenship

4045 CEDARWOOD LANE, LOVELAND, OH. 45140

Residence and/or Mailing Address (the same if only one address is listed, please indicate separately if not the same)

PAGE 12/12 * RCVD AT 3/15/2006 1:35:49 PM [Eastern Standard Time] * SVR:USPTO-EFXRF-2/8 * DNIS:2738300 * CSID:5132486680 * DURATION (mm-ss):05-18

OIPE IAP80
AUG 24 2006
PATENT & TRADEMARK OFFICE

JFW

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
WASHINGTON, D.C., UNITED STATES OF AMERICA

In re Application of:

Applicant(s): CLEVELAND, et al.

Serial No.: 11/348,150

Filing Date: FEBRUARY 6, 2006

Entitled: BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN .....

Examiner:

Our File Ref.: FDS-029388-US

Group Art Unit: 1772

INFORMATION DISCLOSURE STATEMENT
SUBMITTED PERSUANT TO 37 C.F.R. 1.97(b)(1)

Commissioner for Patents
United States Patent and Trademark Office
Post Office Box 1450
Alexandria, Virginia 22313-1450

Dear Commissioner:

In accordance with the duty of disclosure under 37 C.F.R. 1.56, Applicant(s) hereby notifies the U.S. Patent and Trademark Office of certain documents which the Examiner might deem relevant to the issue of patentability of the claims pending in the above-referenced patent application. Enclosed herewith please find Form PTO/SB/08, Information Disclosure Statement by Applicant, 3 sheet(s), and a copy of each reference cited thereon.

This Information Disclosure Statement is filed before the mailing date of a first Office Action on the merits. As such, neither a fee under 37 C.F.R. 1.17(p) nor a statement under 37 C.F.R. 1.97(e) is required; Applicant(s) respectfully requests pursuant to 37 C.F.R. 1.97(b)(3) that the Examiner considers each of the references disclosed herewith.

The submission of the documents enclosed herewith is not intended as an admission that such documents constitute prior art under Title 35 of the United States Code, as against the claims of the above-referenced patent application. Applicant(s) does not waive any right to take

action that would be appropriate to antedate or otherwise overcome the use of any document submitted herewith as prior art under Title 35 of the United States Code.

Applicant(s) respectfully requests that the Examiner initial each of the references listed on the attached Form PTO/SB/08 and return same to the undersigned attorney in accordance with MPEP 609.

Should the Examiner have any questions relating to the submission of the within Information Disclosure Statement or relating to the copies submitted therewith, Applicant(s) respectfully request that the Examiner contact the undersigned counsel, who can be reached at the telephone numbers listed below.

Respectfully submitted,

Date: August 21, 2006

Daniel C. Stelter, Reg. 40,830
Counsel for Applicant
International Paper Company
6285 Tri-Ridge Boulevard
Loveland, Ohio 45140
513.248.6000 telephone
513.248.6455 facsimile

Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
Page 60 of 1179

OIPE IAP60
AUG 24 2006
PATENT & TRADEMARK OFFICE

DANIEL C. STELTER
SENIOR COUNSEL
INTELLECTUAL PROPERTY

INTERNATIONAL PAPER

CINCINNATI TECHNOLOGY CENTER
6285 TRI-RIDGE BOULEVARD
LOVELAND OH 45140

T 513 248 6208
F 513 248 6455
daniel.stelter@ipaper.com

August 21, 2006

Commissioner for Patents
Post Office Box 1450
Alexandria, Virginia 22313-1450

RE: SUBMISSION OF INFORMATION DISCLOSURE STATEMENT

Applicant	:	CLEVELAND, et al.
Serial No.	:	11/348,150
Filed on	:	FEBRUARY 6, 2006
Title	:	BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN.....
Our Ref.	:	FDS-029388-US

Dear Commissioner:

Enclosed herewith for filing, Applicant(s) respectfully submit(s) the following checked items:

- [x] Information Disclosure Statement by Applicant(s), 2 sheet(s), to which Form PTO/SB/08, 3 sheet(s), has been attached along with a copy of each reference(s) cited thereon;
- [ ] Supplemental Information Disclosure Statement by Applicant(s), _____ sheet(s), to which Form PTO/SB/08, _____ sheet(s), has been attached along with a copy of each reference(s) cited thereon; and,
- [x] a postage-prepaid, return-receipt postcard with a listing of the above-checked items.

Please stamp the enclosed postcard and return same to me to indicate your receipt of the above-listed items. Please feel free to contact me if you have any questions concerning the above or the enclosed.

With kindest regards,

Daniel C. Stelter
Attorney for Applicant(s)

Enclosures
DCS/jat

CERTIFICATE OF MAILING OR TRANSMISSION UNDER 37 C.F.R. § 1.8

The undersigned hereby certifies that a true and accurate copy of the documents checked hereon, together with all attachments referred-to herein, is being transmitted to the Honorable Commissioner for Patents, either by first-class mail, postage prepaid, addressed to Commissioner for Patents, Post Office Box 1450, Alexandria, Virginia 22313-1450, or by facsimile transmission to the facsimile number indicated hereon, on this the 21st day of August 20 06.

Jane A. Tomlinson

CIPO IAP60 AUG 24 2006 PATENT & TRADEMARK OFFICE

PTO/SB/08A (08-03)
Approved for use through 07/31/2006. 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.

Substitute for form 1449/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 1 of 3

Complete if Known	
Application Number	11/348,150
Filing Date	FEBRUARY 6, 2006
First Named Inventor	CLEVELAND, et al.
Art Unit	1772
Examiner Name	
Attorney Docket Number	FDS-029388-US

U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
	A	US- 3,901,838	08-26-1975		
	B	US- 5,096,941	03-17-1992		
	C	US- 5,416,133	05-16-1995		
	D	US- 5,458,933	10-17-1995		
	E	US- 5,498,385	03-12-1996		
	F	US- 5,534,616	07-09-1996		
	G	US- 5,565,503	10-15-1996		
	H	US- 5,593,778	01-14-1997		
	I	US- 5,602,227	02-11-1997		
	J	US- 5,618,855	04-08-1997		
	K	US- 5,620,281	04-15-1997		
	L	US- 5,685,756	11-11-1997		
	M	US- 5,693,389	12-02-1997		
	N	US- 5,854,304	12-29-1998		
	O	US- 5,939,467	08-17-1999		
	P	US- 6,033,747	03-07-2000		
	Q	US- 6,040,063	03-21-2000		
	R	US- 6,080,478	06-27-2000		
	S	US- 6,096,810	08-01-2000		

FOREIGN PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Foreign Patent Document Country Code[3]-Number[4]-Kind Code[5] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages Or Relevant Figures Appear	T[6]
	F1	JP2002-322307	11-08-2002			
	F2	JP2003-276144	09-30-2003			
	F3	JP2003-41142	02-13-2003			

Examiner Signature		Date Considered	

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant. [1]Applicant's unique citation designation number (optional). [2]See Kinds Codes of USPTO Patent Documents at www.uspto.gov or MPEP 901.04. [3]Enter Office that issued the document, by the two-letter code (WIPO Standard ST.3). [4]For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. [5]Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. [6]Applicant is to place a check mark here if English language Translation is attached.

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 2 hours 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, 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 (1-800-786-9199) and select option 2.

PTO/SB/08A (08-03)
Approved for use through 07/31/2006. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
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Substitute for form 1449/PTO

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Sheet 2 of 3

Complete if Known	
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Filing Date	FEBRUARY 6, 2006
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Art Unit	1772
Examiner Name	
Attorney Docket Number	FDS-029388-US

U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
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Complete if Known	
Application Number	11/348,150
Filing Date	FEBRUARY 6, 2006
First Named Inventor	CLEVELAND, et al.
Art Unit	1772
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U. S. PATENT DOCUMENTS

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PATENT ABSTRACTS OF JAPAN

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C08L 1/12
C08L 67/00

(21)Application number : 2001-163457
(22)Date of filing : 23.04.2001

(71)Applicant : PLUSTO:KK
(72)Inventor : HAYASHIBARA HITOSHI

(54) BIODEGRADABLE RESIN FOAM FOR PACKAGING

(57)Abstract:
PROBLEM TO BE SOLVED: To contribute to solve waste problems by developing a biodegradable resin foam sheet for packaging capable of replacing a formed polystyrene sheet used for packaging fruits, or the like.
SOLUTION: This biodegradable resin foam mainly comprises 35-90 wt.% of a cellulose acetate, 5-60 wt.% of a biodegradable co-polyester, 2-20 wt.% of a non-wood plant fiber powder, 2-20 wt.% of a bamboo-trunk fiber powder treated at a high temperature and 1-10 wt.% of a low polymerization degree polyethylene glycol. The biodegradable resin foam for packaging applied with a natural anti-bacterial agent on the surface is also provided.

LEGAL STATUS

[Date of request for examination]

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[Patent number]

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[Date of requesting appeal against examiner's decision of rejection]

[Date of extinction of right]

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(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2002－322307
（P2002－322307A）
(43)公開日　平成14年11月８日(2002.11.8)

(51)Int.Cl.'	識別記号	ＦＩ		テーマコード'(参考)
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Ｂ６５Ｄ　65/46		Ｂ６５Ｄ　65/46		３Ｅ０６７
81/107		81/28	Ｃ	３Ｅ０８６
81/28		85/34	Ｇ	３Ｅ０９６
85/34		Ｃ０８Ｊ　9/36		４Ｆ０７４

審査請求　未請求　請求項の数２　書面　（全　４　頁）　最終頁に続く

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(71)出願人　596035097
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弁理士　竹田　逸郎

最終頁に続く

(54)【発明の名称】　包装用生分解性樹脂発泡体

(57)【要約】
【課題】果物等を包装するため使用されている発泡ポリスチレンシートに代替しうる包装用生分解性樹脂発泡シートを開発し、廃棄物問題の解決に寄与する。
【解決手段】主として、セルロースアセテート３５～９０重量%、生分解性コポリエステル５～６０重量%、非木材質植物繊維粉末２～２０重量%、高温処理した竹の幹の繊維粉末２～２０重量%、及び、低重合度ポリエチレングリコール１～１０重量%からなることを特徴とする包装用生分解性樹脂発泡体、ならびに、その表面に天然の抗菌剤を塗布したことを特徴とする包装用生分解性樹脂発泡体を提供する。

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【特許請求の範囲】
【請求項１】　主として、セルロースアセテート３５～９０重量%、生分解性コポリエステル５～６０重量%、非木材質植物繊維粉末２～２０重量%、高温処理した竹の幹の繊維粉末２～２０重量%、及び、低重合度ポリエチレングリコール１～１０重量%からなることを特徴とする包装用生分解性樹脂発泡体
【請求項２】　請求項１記載の包装用生分解性樹脂発泡体の表面に、更に天然の抗菌剤を塗布したことを特徴とする包装用生分解性樹脂発泡体
【発明の詳細な説明】
【０００１】
【発明の属する技術分野】本発明は、主として、梨、りんご、洋梨等の果物を包むために使用される発泡ポリスチレンシートや、それを収納するプラスチックケースや、ダンボール箱の内装に使用される緩衝シート等に代替しうる包装用生分解樹性脂発泡体に関するものである。
【０００２】
【従来の技術】従来、梨、りんご、洋梨等の果物は、和紙や藁のついた紙で包んで木綿等の緩衝材が充填された箱の中に入れて輸送されてきたが、最近になって、これらの果物は、薄手の発泡ポリスチレンシートで包んでから、プラスチックケースに詰められ、それをダンボール箱に入れて、上から発泡ポリスチレンシート等で覆い、蓋をしてダンボール箱に収納して輸送することが多くなった。
【０００３】
【発明が解決しようとする課題】前述のように、果物等を包むために薄手の発泡ポリスチレンシートや、プラスチックケースや、緩衝用の発泡ポリスチレンシートは、使用後に廃棄し処理するに当たって、嵩高のため、収集し運搬するにも、また、投棄場に投棄するにも大きな容積を占め、また、焼却するにしても高温になり炉を損傷する等の問題があった。そのため、多種多様な生分解性樹脂や生崩壊性樹脂が開発されてきた。しかしながら、これらの生分解樹脂又は生崩壊樹脂は、ポリスチレンの異なり、予備発泡させた後、型に入れて本発泡させて成型品を作ることが困難であった。そのため多くの技術が開発されてきた。
【０００４】生分解性樹脂発泡体の成型品を製造する方法には、小発泡体を接着させて成型品を製造する方法と成型品の形状に発泡させる方法がある。小発泡体を接着させる方法には、本出願人が特開平１０－３３０５２７号公報で開示した方法や特開２０００－１４１５００号公報で開示した方法等があり、所定の形状に発泡させる方法には、本出願人が特願平２０００－３０２９１０号公報で開示した方法等がある。この公報に開示した方法は、押出機のダイに隣接して設けられた所定の断面形状の外周に内接する成型枠の中で発泡させるものであり、包装用に使用されるシート状の生分解樹脂発泡成型品も製造することができる。しかしながら、得られたシート状の製品は、長手方向には高い引張強度を有しているが、横方向には、長手方向の引張強度よりかなり低く、引き裂け易いという欠点があった。なお、この問題を解決するため、フィルムを製造するように、Ｔダイから樹脂を押し出して発泡させて発泡生分解樹脂シートを製造したところ、確かに、横方向の引張強度が高くなったが、必ずしも充分なものではなかった。
【０００５】このような問題を解決するため、本発明者は、特願２０００－２４３４１９号明細書で開示したように、セルロースアセテートに、引張強度を上げるため非木材質繊維を混入し、更に、柔軟性を上げるためポリエチレングリコールを添加して、それなりの引張強度を有するシート状の生分解性樹脂発泡体を製造することができた。しかしながら、果物等の包装用シートとしては、必ずしも、引張強度や柔軟性の点で充分なものではなかった。本発明が解決しようとする課題は、更に引張強度や柔軟性を高め、果物の鮮度保持性も付加して、果物等を包んだり、果物等のケースにしたり、果物等の緩衝材としたりすることができる包装用の生分解性樹脂発泡成型体を提供することにある。
【０００６】
【課題を解決するための手段】本発明者は、前述の課題を解決するために、主として、セルロースアセテート３５～９０重量%、生分解性コポリエステル５～６０重量%、非木材質植物繊維粉末２～２０重量%、高温処理した竹の幹の繊維粉末２～２０重量%、及び、低重合度ポリエチレングリコール１～１０重量%からなることを特徴とする包装用生分解性樹脂発泡体（以下「第１発明」という）、ならびに、第１発明に係わる包装用生分解性樹脂発泡体の表面に、更に天然の抗菌剤を塗布したことを特徴とする包装用生分解性樹脂発泡体（以下「第２発明」という）を提供する。
【０００７】第１発明におけるセルロースアセテートは、セルロースの水酸基を無水酢酸等でアセチル化したもので、酢化の程度によって各種のものがある。このセルロースアセテートは、発泡体の骨格をなすものであり、このような目的に使用できるものであれば、どのようなセルロースアセテートでもよい。通常は、市販のセルロースアセテートが、そのまま使用される。生分解性コポリエステルは、脂肪鎖と芳香環を有するポリエステルであって生分解性を有するものある。この生分解性コポリエステルは、骨格となるセルロースアセテートの補強剤として作用し、非木材質植物繊維粉末、及び、高温処理した竹の幹の繊維粉末の接着剤として作用するものと考えられる。現在、イーストマンケミカル社の「イースターバイオ」やＢＡＳＦ社の「エコフレックス」等が知られている。また、生分解性コポリエステルの中には、「イースターバイオ」のように、１００℃以上の高

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い融点を有するものもある。

【０００８】このセルロースアセテートと生分解性コポリエステルとの比率は、セルロースアセテートが３５～９０重量%、生分解性コポリエステルが５～６０重量%の範囲であり、生分解性コポリエステルが、これより少ないと、補強効果や接着効果が現れず、また、これより多くしても、これれらの効果がそれに比例して増強されなくなる。また、コストも高くなる。

【０００９】また、非木材質植物繊維粉末は、草本類をパルプ化したものであり、（株）チャフローズコーポレーション製「チャフローズ」が知られている。この非木材質植物繊維粉末を添加することにより、横方向の引張強度と柔軟性とを高めることが期待される。高温処理した竹の幹の繊維粉末は、皮を剥いだ竹の幹を高温処理して粉末化した繊維粉末（以下「竹鮮」という）であり、（株）タケックス製「パデオール」が知られている。この竹鮮を添加することにより、横方向の弾力性と引張強度を高め、果物等の被包装物の鮮度保持性を持たせることが期待される。

【００１０】これらの非木材質植物繊維粉末及び竹鮮の添加比率は、非木材質植物繊維粉末が２～２０重量%、竹鮮が２～２０重量%の範囲であり、これらの範囲未満であれば、縦方向及び横方向の引張強度を高める効果が現れず、また、これらの範囲を超えても、それに比例して引張強度が高くならなくなる。また、シート状に成型するのも困難になる。

【００１１】また、低重合度ポリエチレングリコールは、医薬品や化粧品に使用されているものであり、通常ポリエチレングリコール４００が用いられる。この低重合度ポリエチレングリコールは発泡体に弾性や柔軟性を与えるものである。また、その添加比率は１～１０重量%の範囲で充分である。その他に添加されるものには、ほたて貝殻を高熱処理して得られた抗菌性、天然色素、薬草粉末、薬草を煎じた液、天然の顔料等があげられる。

【００１２】第２発明に係わる包装用生分解性樹脂発泡体は、第１発明に係わる包装用生分解性樹脂発泡体の表面に、更に天然の抗菌剤を塗布したことを特徴としている。この天然の抗菌剤には、穀物から得られた天然フラボノイド系抗菌剤として、ニッセキ（株）製「セントリスＥ－１」等が知られている。

【００１３】第１発明に係わる包装用生分解樹脂発泡体を製造するには、主として、セルロースアセテート３５～９０重量%に水を添加して膨潤させ、非木材質植物繊維粉末２～２０重量%、竹鮮２～２０重量%、低重合度ポリエチレングリコール１～１０重量%、及び、生分解性コポリエステル１５～３０重量%を添加して、エクストルーダーに入れ、加熱溶融してＴダイから押し出し、断面が幅の広い矩形状になっている筒状の成形枠の中で発泡させて、シート状の成形品にする方法がとられる。

【００１４】第２発明に係わる包装用生分解性樹脂発泡体を製造するには、前述の方法で得られたシート状の成型品の両面又は片面に、天然の抗菌剤の水溶液を噴霧する等の方法により、天然の抗菌剤を塗布する方法がとられる。

【００１５】

【発明の実施の態様】１．第１発明に係わる包装用生分解樹性脂発泡体等の製造

〔実施例１〕セルロースアセテート（帝人（株）製「アセテートフレーク」）６．７ｋｇに水０．７ｋｇを添加して膨潤させ、非木材質植物繊維粉末（（株）チャフローズコーポレーション製「チャフローズ」）０．３ｋｇ、竹鮮（（株）タケックス製「パデオール」）０．５ｋｇ、ポリエチレングリコール（４００）０．５ｋｇ、及び、生分解性コポリエステル（イーストマンケミカル社製「イースターバイオＧＰ」）２．０ｋｇを加えて混合し、二軸押出機（φ４６ｍｍ、Ｌ／Ｄ２１．５）に供給し、シリンダー温度$C_1$８５℃、$C_2$１２４℃、$C_3$１３１℃、$C_4$１４０℃、$C_5$１９４℃、$C_6$１８１℃、$D_1$１７１℃、$D_2$１８０℃で、幅１００ｃｍ、スリット間隔１ｍｍのノズルから押し出し、発泡させて、幅１００ｃｍ、厚さ２．０ｍｍの第１発明に係わる包装用生分解性樹脂発泡体（以下「シートＡ」という）を製造した。

【００１６】〔比較例〕セルロースアセテート（帝人（株）製「アセテートフレーク」）７．８ｋｇに水０．７ｋｇを添加して膨潤させ、非木材質植物繊維粉末（（株）チャフローズコーポレーション製「チャフローズ」）０．８ｋｇ、および、ポリエチレングリコール（４００）２．０ｋｇとを加えて混合し、二軸押出機（φ４６ｍｍ、Ｌ／Ｄ２１．５）に供給し、シリンダー温度$C_1$７０℃、$C_2$９０℃、$C_3$１３０℃、$C_4$２００℃、$C_5$２１０℃、Ｄ１９０℃で、幅１００ｃｍ、スリット間隔１ｍｍのノズルから押し出し、発泡させて、幅１００ｃｍ、厚さ２．０ｍｍの比較のための包装用生分解性樹脂発泡体（以下「シートＢ」という）を製造した。

【００１７】２．第２発明に係わる包装用生分解樹性脂発泡体の製造

〔実施例２〕実施例１で得られたシートＡ」の両面に、天然フラボノイド系抗菌剤（ニッセキ（株）製「セントリスＥ－１」）２．０重量%水溶液を噴霧して、第２発明に係わる包装用生分解樹性脂発泡体（以下「シートＣ」という）を製造した。

【００１８】３．引張強度等の特性の比較

実施例１で得られたシートＡは、比較例で得られたシートＢに比べて、ポリエチレングリコールの添加量が少ないにもかかわらず、表面が滑らかで、柔軟性があった。また、シートＡとシートＢとのそれぞれについて、１０個の縦１２０ｍｍ、横１２０ｍｍ（厚さ２．０ｍｍ）の

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サンプルを作成して、それぞれ、その中の５個について、縦方向の一端を固定し他端をばね秤を介して引っ張り、切断した時のばね秤の表示を縦方向の引張強度として測定した。また、それぞれ残り５個について、横方向の一端を固定して、同様にして横方向の引張強度を測定した。その結果、縦方向の平均の引張強度は、従来品のシートＢは３．５ｋｇであるのに対して、本発明に係わるシートＡは５．５ｋｇに向上していた。また、横方向の平均の引張強度は、従来品のシートＢは１．８ｋｇであるのに対して、本発明に係わるシートＡは２．０ｋｇに向上していた。

【００１９】このように、縦方向の引張強度も横方向の引張強度が向上したのは、生分解性コポリエステルが、添加された非木材質植物繊維粉末や高温処理した竹の幹の繊維粉末を、セルロースアセテート発泡体中に接着する作用を果たしているためと考えられる。また、縦方向の引張強度が大きく向上するのに対して、横方向の引張強度の向上が僅かであるのは、添加された非木材質植物繊維粉末や高温処理した竹の幹の繊維粉末が、Ｔダイから押し出される際に流れ方向に配向する傾向にあって、それが発泡の際にもあまり乱されないためと考えられる。

【００２０】４．鮮度保持試験

実施例２で得られた第２発明に係わるシートＣ１０枚、及び、従来品のシートＢ１０枚で、それぞれ、１０個の林檎を包み、１週間室温で保管したところ、従来品のシートＢで包んだ林檎には黄味を帯びていたが、シートＣで包んだ林檎には全く変化が見られなかった。

【００２１】

【発明の効果】本発明に係わる包装用生分解性樹脂発泡体は、前述のような構成であって、包装材料に適した引張強度を有し、かつ、果物等に対する鮮度保持作用を有するので、これまで果物の包装に使用されてきた発泡ポリスチレンシートやプラスチックケースに代替して、果物等の包装等に幅広く使用することができ、プラスチック廃棄物に関する問題の解決に寄与し、環境の保全に貢献し、国民の生活の向上に大きく貢献するものである。

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(54) LAMINATED FILM OF BIODEGRADABLE POLYESTER

(57)Abstract:
PROBLEM TO BE SOLVED: To provide a laminated film of a biodegradable polyester which has improved moistureproofness without marring biodegradability.
SOLUTION: The laminated film of the biodegradable polyester has a low molecular weight polyolefin layer preferably falling within the viscosity-average molecular weight range of 1,000 to 7,000, laminated on at least one side of a base material layer of a biodegradable polyester film comprising an aliphatic polyester such as polylactic acid or the like and also improved moistureproofness without marring biodegradability.

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最終頁に続く

(54)【発明の名称】　生分解性ポリエステル積層フィルム

(57)【要約】
【課題】生分解性を損なうことなく、防湿性が改良された生分解性ポリエステル積層フィルムを提供する。
【解決手段】ポリ乳酸等の脂肪族ポリエステルからなる生分解性ポリエステルフィルム基材層の少なくとも片面に好ましくは粘度平均分子量が１０００～７０００の範囲にある低分子量ポリオレフィン層が積層されてなる、生分解性を損なうことなく、防湿性が改良された生分解性ポリエステル積層フィルムに関する。

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【特許請求の範囲】
【請求項１】生分解性ポリエステルフィルムからなる基材層の少なくとも片面に低分子量ポリオレフィン層が積層されてなることを特徴とする生分解性ポリエステル積層フィルム。
【請求項２】生分解性ポリエステルフィルムが、二軸延伸されてなる請求項１記載の生分解性ポリエステル積層フィルム。
【請求項３】低分子量ポリオレフィンが、粘度平均分子量が１０００～７０００の範囲にある請求項１記載の生分解性ポリエステル積層フィルム。
【請求項４】低分子量ポリオレフィンが、低分子量エチレン系重合体である請求項１又は３記載の生分解性ポリエステル積層フィルム。
【請求項５】生分解性ポリエステルが、ポリ乳酸である請求項１記載の生分解性ポリエステル積層フィルム。
【請求項６】生分解性ポリエステルが、芳香族系ポリエステルである請求項１記載の生分解性ポリエステル積層フィルム。
【請求項７】芳香族系ポリエステルがスルホン酸金属塩基を核置換基として有する芳香族ジカルボン酸を含む芳香族系ポリエステルである請求項６記載の生分解性ポリエステル積層フィルム。
【発明の詳細な説明】
【０００１】
【産業上の利用分野】本発明は、防湿性が改良された生分解性ポリエステル積層フィルムに関する。
【０００２】
【従来の技術】生分解性フィルムは、土壌中や水中で加水分解や生分解を受け、徐々に崩壊や分解が進み、最後には微生物の作用で分解物へと変化するものである。そのようなフィルムとして、ポリ乳酸等の脂肪族系ポリエステル樹脂や芳香族系ポリエステル樹脂、ポリビニルアルコール、酢酸セルロース、デンプン等から成形したフィルムが知られており、中でも、ポリ乳酸を用いた二軸延伸フィルムは、包装用フィルムとして大量に用いられているニ軸延伸ポリプロピレンフィルムに似て、透明性、剛性に優れることから、包装用フィルムとして一部で使用されている。しかしながら、かかるポリ乳酸二軸延伸フィルムは、ニ軸延伸ポリプロピレンフィルムに比べると防湿性（水蒸気バリヤー性）に劣ることから包装用フィルムとしての利用範囲が限られている。
【０００３】生分解性ポリエステルフィルムの防湿性を改良する方法として、ポリ乳酸を主成分とするフィルムと再生セルロースフィルムとからなるフィルムが提案されている（特開平５－３８７８４号公報）が、再生セルロース自体がポリオレフィン等に比べ防湿性に劣ることから、得られるフィルムは未だ防湿性が不十分である。
【０００４】
【発明が解決しようとする課題】そこで本発明は、ポリ乳酸等の脂肪族系ポリエステル樹脂や芳香族系ポリエステル樹脂等の生分解性ポリエステルフィルムが本来有する生分解性を損なうことなく、防湿性に優れた二軸延伸フィルムを開発することを目的とした。
【０００５】
【課題を解決するための手段】
【発明の概要】本発明は、生分解性ポリエステルフィルムからなる基材層の少なくとも片面に低分子量ポリオレフィン層が積層されてなることを特徴とする防湿性に優れた生分解性ポリエステル積層フィルムを提供することにある。そして、好ましくは、生分解性ポリエステルとして、ポリ乳酸及び芳香族ポリエステル、特にスルホン酸金属塩基を核置換基として有する芳香族ジカルボン酸を含む芳香族系ポリエステルから選択され、低分子量ポリオレフィンとして粘度平均分子量が１０００～７０００の範囲にあり、又、低分子量ポリオレフィンが低分子量エチレン系重合体である生分解性ポリエステル積層フィルムを提供することにある。
【０００６】
【発明の具体的な説明】生分解性ポリエステル
本発明の基材層を構成する生分解性ポリエステルフィルムの原料である生分解性ポリエステルは、種々公知の生分解性ポリエステルであり、具体的にはポリ乳酸、あるいはコハク酸、グルタル酸、アジピン酸、ピメリン酸、スベリン酸、アゼライン酸、セバシン酸、ウンデカン二酸、ドデカン二酸、シクロペンタンジカルボン酸およびシクロヘキサンジカルボン酸等の通常、炭素数が４～１２の二塩基性カルボン酸とエチレングリコール、トリエチレングリコール、１，４－ブタンジオール、ペンタメチレングリコール、１，８－オクチレングリコール、ナノメチレングリコール、デカメチレングリコール等の通常、炭素数が２～１０のグリコールから任意に選ばれた１種あるいは２種以上のジカルボン酸とグリコールの脱水縮合あるいはそれに続く脱グリコール反応により得られる生分解性を有する脂肪族ポリエステル、あるいは芳香族ジカルボン酸、脂肪族グリコール、およびスルホン酸金属塩基を核置換基として有する芳香族ジカルボン酸からなる生分解性を有する芳香族系ポリエステルである。これらの中でも、融点が１５０℃以上のポリ乳酸及び芳香族ポリエステル、特にスルホン酸金属塩基を核置換基として有する芳香族ジカルボン酸を含む芳香族系ポリエステルが、防湿性能、酸素バリア性能、透明性、強度、柔軟性といったフィルム物性に優れるので好ましい。
【０００７】ポリ乳酸
生分解性ポリエステルの例である本発明に係わるポリ乳酸は、乳酸の構造単位がＬ－乳酸であるポリ（Ｌ－乳酸）、構造単位がＤ－乳酸であるポリ（Ｄ－乳酸）、更にはＬ－乳酸とＤ－乳酸の混合物（ラセミ体）であるポリ（ＤＬ－乳酸）等の乳酸の単独重合体、及び乳酸を主

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成分とし、乳酸以外の共重合可能なコモノマーを少割合、例えばグリコール酸５０重量%未満、好ましくは３０重量%以下、より好ましくは１０重量%以下の割合で共重合したコポリマー、あるいはこれらの混合物等の乳酸を主成分とする重合体である。乳酸と共重合可能なコモノマーとしては、例えば３－ヒドロキシブチレート、カプロラクトン、グリコール酸などを挙げることができる。これらの中でも乳酸のみからなる単独重合体は透明性に優れているので好ましく使用できる。かかるポリ乳酸の重合法としては、縮重合法、開環重合法など公知のいずれの方法を採用することができる。例えば、縮重合法ではＬ－乳酸またはＤ－乳酸あるいはこれらの混合物を直接脱水縮重合して任意の組成を持ったポリ乳酸を得ることができる。

【０００８】また、開環重合法では乳酸の環状２量体であるラクチドを、必要に応じて重合調整剤等を用いながら、触媒を使用してポリ乳酸を得ることができる。ラクチドにはＬ－乳酸の２量体であるＬ－ラクチド、Ｄ－乳酸の２量体であるＤ－ラクチド、さらにＬ－乳酸とＤ－乳酸からなるＤＬ－ラクチドがあり、これらを必要に応じて混合して重合することにより任意の組成、結晶性を有するポリ乳酸を得ることができる。

【０００９】ポリ乳酸の分子量増大を目的として少量の鎖延長剤、例えば、ジイソシアネート化合物、エポキシ化合物、酸無水物などを使用できる。ポリ乳酸の重量平均分子量は通常６万～１００万の範囲にあり、この範囲を下回る場合は用途によっては実用物性がほとんど発現されない虞があり、一方、上回る場合には、溶融粘度が高すぎ成形加工性に劣る虞がある。

【００１０】芳香族ポリエステル

生分解性ポリエステルの他の例である本発明に係わる芳香族ポリエステルは、生分解性を有するものであり、好ましくはスルホン酸金属塩基を核置換基として有する芳香族ジカルボン酸を１共重合成分として含むポリエステルであり、より具体的には、芳香族ジカルボン酸、脂肪族グリコール、およびスルホン酸金属塩基を核置換基として有する芳香族ジカルボン酸、それに必要に応じて脂肪族ジカルボン酸または脂肪族ヒドロキシカルボン酸を加え、それらの成分間で重縮合反応を行って得られるポリエステルであることが望ましい。芳香族ポリエステルの好ましい組成は、芳香族ジカルボン酸成分に由来する単位が３０～４９．９モル%、脂肪族グリコール成分に由来する単位が３５～５０モル%、スルホン酸金属塩基を置換基として有する芳香族または脂肪族ジカルボン酸成分に由来する単位が０．１～５モル%、および脂肪族ジカルボン酸または脂肪族ヒドロキシカルボン酸成分に由来する単位が０～３０モル%（ここで、全単位の合計が１００モル%になる）である。また得られるフィルムに可撓性、生分解性等の性能を付与し、また向上させるために、さらに脂肪族ジカルボン酸または脂肪族ヒドロキシカルボン酸を共重合成分として加えた多成分系のポリエステルであってもよい。そのような芳香族ポリエステルは、特表平５－５０７１０９号公報、特表平６－５０５０４０号公報、特表平６－５０５５１３号公報等に詳しい。

【００１１】本発明に係わる芳香族ポリエステルは、その重量平均分子量が、１０，０００～５００，０００の範囲が好ましい。また、そのメルトフローレートは、ＡＳＴＭ Ｄ－１２３８に準拠し、２２０℃、２１６０ｇ荷重下で測定した値が、０．１～１００（ｇ／１０分）であることが好ましい。分子量およびメルトフローレートが前記の範囲内にあると、押出成形に適した溶融粘度を示し、また二軸延伸フィルムとしての十分な機械的強度を有する。

【００１２】低分子量ポリオレフィン

本発明の生分解性ポリエステルフィルムからなる基材層の少なくとも片面に積層される低分子量ポリオレフィン層となる低分子量ポリオレフィンは、エチレン、プロピレン、１－ブテン、１－ヘキセン、１－オクテン、４－メチル－１ペンテン等の炭素数２から１０のα－オレフィンの単独若しくは共重合体であり、通常、粘度平均分子量が１０００～７０００、好ましくは１０００～６０００の範囲にある。粘度平均分子量が１０００未満のものは、基材層に積層して得られる積層フィルムの耐摩耗性、塗膜強度等に劣る虞があり、一方、７０００を越えるものは、得られる積層フィルムの生分解性が低下する傾向にある。また、これら低分子量ポリオレフィンは、種々公知の方法、例えば、溶融状態にあるワックスを、約１２０乃至２００℃、好ましくは約１４０乃至１７０℃で酸素含有ガスと接触させ、酸化した酸化物であっても、あるいは、低分子量ポリオレフィンにマレイン酸、無水マレイン酸等の不飽和カルボン酸またはその誘導体成分をグラフト共重合してなるグラフト変性物であってもよい。

【００１３】これら、低分子量オレフィン重合体の中でも、エチレンの単独重合体あるいはエチレンを主体とした重合体等のエチレン系重合体が好ましく、特に融点が９０～１３５℃、より好ましくは１１０～１３５℃の範囲にあり、且つ、密度が０．９０～０．９５ｇ／ｃｍ^3、より好ましくは０．９１～０．９４ｇ／ｃｍ^{3}の範囲にあるものが防湿性に優れる。またかかる低分子量ポリオレフィンには得られる積層フィルムの防湿性能をより向上させる目的で粒径０．１～５μｍのヘクトライト等の無機層状化合物を配合してもよい。また、得られる積層フィルムの生分解速度を向上させる目的で他の各種生分解性のプラスチックを配合してもよい。

【００１４】生分解性ポリエステル積層フィルム

本発明の生分解性ポリエステル積層フィルムは、生分解性ポリエステルフィルムからなる基材層の少なくとも片面に低分子量ポリオレフィン層が積層されてなる。基材

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層及び低分子量ポリオレフィン層の厚さは用途に応じて種々決定されるが、通常、基材層の厚さが、２０～１００μｍ、好ましくは２５～５０μｍ、低分子量ポリオレフィン層の厚さが、０．１～５μｍ、好ましくは０．５～３μｍの範囲にある。

【００１５】本発明の生分解性ポリエステル積層フィルムは種々公知の方法で得られるが、低分子量ポリオレフィンは溶融粘度が低いので、通常は、予め製造した生分解性ポリエステルフィルムの少なくとも片面に、低分子量ポリオレフィンの水性分散体を被覆して乾燥する方法が好ましい。基材層となる生分解性ポリエステルフィルムは無延伸フィルムでも、一軸あるいは二軸延伸フィルムでもよいが、二軸延伸フィルムが得られる積層フィルムの機械的強度、防湿性等に優れるので好ましい。

【００１６】低分子量ポリオレフィンの水性分散体としては、通常、粒子径が０．０１～１０μｍ、好ましくは０．１～９μｍの低分子量ポリオレフィン粒子を水に分散した分散体である。又、基材層に被覆（塗布）した後、乾燥を速やかに行えるようにアルコールを０～５０溶液重量%、好ましくは１０～３０溶液重量%含む水性分散体にしておいても良い。低分子量ポリオレフォンの水性分散体には生分解性ポリエステルフィルムとの接着性若しくは濡れ性改良のために水系イソシアネート系接着材、セロソルブ系等の有機溶剤、粘度調整剤、防かび剤、静防剤などを添加することができる。水性分散体の濃度は、コーティングに支障のない範囲で高い方が良い。通常１０～５０%、好ましくは２０～４０%である。最適な粘度は塗布方法や製造速度により異なるが、例えば、グラビアロールコーティングにおいては２００ｃｐｓ以下が好ましい。

【００１７】低分子量ポリオレフィン水性分散体の塗布方法は、従来使用されているものが利用できる。例えば、ダイレクトグラビアコーティング、グラビアオフセットコーティング、アークグラビアコーティング、グラビアリバースコーティング等のグラビアコーティング、トップフィードリバースロールコーティング、ボトムフィードリバースロールコーティング、ノズルフィードリバースロールコーティング等のリバースロールコーティング、キスロールコーテイング、マイヤーバーコーティング、ドクターブレードコーティング、及びエアーナイフコーティングなどが挙げられる。これらの中でもグラビアコーティング、特にグラビアリバースコーティングが好ましい。

【００１８】生分解性ポリエステルフィルムと低分子量ポリオレフィンとの接着性、耐水性をより改良するために、基材層の表面に下塗り剤としてイソシアネート系などの接着剤を生分解性ポリエステルフィルムに塗布しておいた方が好ましい。かかる下塗り剤の塗布量は、通常、固形分重量で５０～１５０ｍｇ／ｍ^{2}の範囲が好ましい。前記被覆用組成物の塗布量は、通常、固形分重量で０．１～５ｇ／ｍ^2、好ましくは０．５～２ｇ／ｍ^{2}の範囲にある。また上記下塗り剤の濡れを良くするため０．１～２５Ｗ／ｍ^2・分のコロナ放電処理を行うことが好ましい。

【００１９】低分子量ポリオレフィンの水性分散体を塗布した生分解性ポリエステル積層フィルムの乾燥温度は、通常、５０～１２０℃、好ましくは６０～９０℃の範囲がよい。乾燥温度が低いと、良好な被膜が形成されなかったり、乾燥時間を長くすることを必要とし、生産性が低下する虞がある。一方、乾燥温度が高すぎると、代表的な生分解性ポリエステルフィルムであるポリ乳酸等の耐熱性が低いフィルムは乾燥時に軟化してシワが発生する虞がある。また低分子量ポリオレフィンを融解し防湿膜とするには乾燥と同時又は乾燥後に１２０～１６０℃の熱処理を行う必要がある。かかる熱処理は生分解性ポリエステルフィルムに負荷を与えるため、オーブン等で端部を固定しながら行うのが望ましく、また紙またはデンプンシートにラミネートした後に容器成形の金型の熱で成形と同時に熱処理を行っても良い。

【００２０】

【発明の効果】本発明の生分解性ポリエステル積層フィルムは、高い機械的強度を有し、かつ優れた水蒸気ガスバリヤー性を有するので、高い水蒸気バリア性能が必要なため、従来ポリオレフィンフィルムしか使用できなかった乾物等の食品包装も可能となり、内容物（食品）も含めたコンポストごみの回収、処理が容易になり、生分解性プラスチック製品の用途を大きく広げることができる。その他発明の利用形態として、紙とのラミを用いた食品または廃棄用のコンポスト容器等が挙げられる。

又、かかる生分解性ポリエステル積層フィルムを紙、パルプモールドまたはデンプン混練樹脂に積層して包装材料、トレー、容器等として利用することもできる。

【００２１】

【実施例】次に実施例を挙げて本発明を更に具体的に説明するが、本発明はその要旨を越えない限りこれらの実施例に制約されるものではない。

【００２２】実施例及び比較例で得られた積層体の物性値は以下の方法で測定した。

（１）光学特性

日本電色工業社製　ヘイズメーター３００Ａを用いてヘイズ、平行光線透過率（ＰＴ）及びグロスを測定した。

（２）透湿度

ＪＩＳ　Ｚ０２０８　防湿包装材料の透湿度試験（カップ法）による。

【００２３】実施例及び比較例で使用した原料は次の通りである。

（１）低分子量ポリエチレンの水性分散体（ＰＯ－１）

低分子量ポリエチレンの物性：密度：０．９２ｇ／ｃｍ^3、針入度法硬度（ＪＩＳ　Ｋ２２０７）：３、融点：１１０℃、粘度平均分子量：４０００水性分散体の物

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性：固形分濃度：４０重量%、ｐＨ：９、粘度：１００ＯＣＰ、粒径：４μｍ

（２）低分子量ポリエチレンの水性分散体（ＰＯ－２）

低分子量ポリエチレンの物性：密度：０．９５ｇ／ｃｍ^3、融点：１３２℃、粘度平均分子量：４０００

水性分散体の物性：固形分濃度：４０重量%、ｐＨ：９、粘度：１０００ＣＰ、粒径：７μｍ

（３）生分解性ポリエステル：以下の組成からなるスルホン酸金属塩基を有する芳香族ジカルボン酸を共重合体として含む生分解性ポリエステル樹脂を用いた。共重合体のモノマー組成：テレフタル酸４５モル%、エチレングリコール３７モル%、ジエチレングリコール９モル%、５－スルホーイソフタル酸ナトリウム１モル%、ヒドロキシ酢酸８モル%。共重合体の密度：１．３５（ｇ／ｃｍ^3）、融点：２００℃、メルトフローレート（２２０℃、２１６０ｇ荷重）：１５（ｇ／１０分）。

【００２４】生分解性ポリエステル二軸延伸フィルムの製造：ブルックナー社の逐次二軸延伸機を用いて製膜した。上記生分解性ポリエステル樹脂をオーブン中で予備乾燥し、押出機（６０ｍｍφ、シリンダー設定温度：２３０℃）中へ供給して溶融した。押出機先端に設けたＴダイからフィルム状に押出し、キャストロールで３０℃に冷却し、厚さ２５０μｍのシートを成形した。次に、逐次二軸延伸工程で、このシートを６０℃で３．０倍縦延伸し、次いで６０℃で３．２倍横延伸し、引き続き１８０℃で２０秒間熱処理（ヒートセット）を行った。その後、フィルム両面にコロナ放電処理を施し、厚さ２５μｍの生分解性ポリエステル二軸延伸フィルムを得た。

【００２５】実施例１

上記生分解性ポリエステル二軸延伸フィルムの片面にイソシアネート系接着剤（三井武田ケミカル株式会社製タケネートＡ－３：１ｇに同Ａ－３１０：１２ｇを酢酸エチル１８７ｇで希釈し固形分３．４%液にする）を、マイヤーバーを用いて固形分重量で１００ ｍｇ／ｍ^{2}塗布し、６０℃で１０秒間乾燥した。次いで、前記低分子量ポエチレン水性分散体（ＰＯ－１）をマイヤーバーを用いて固形分重量で ２．０ｇ／ｍ^{2}になるように塗布した後、ドライヤーを用いて７０℃で２０秒間乾燥し積層フィルムを得た。尚、それぞれの塗布液は界面活性剤として花王株式会社製ラウリルエーテルＰＯＥ（１３，３）商品名エマルゲン１２０を０．４ｗｔ%加えてある。得られた積層フィルムを厚み３ｍｍのガラス板に固定し四方を４ｃｍ間隔でクリップで固定し、熱による収縮を抑えて強制対流型高温槽（イズミ製作所製オーブン）に入れ、１６０℃×５分の条件で熱固定処理を行った。得られた積層フィルムの物性値を表１に示す。

【００２６】実施例２

実施例１で用いたは低分子量ポリエチレン水性分散体（ＰＯ－１）に代えて、低分子量ポリエチレン水性分散体（ＰＯ－２）を用いる以外は実施例１と同様に行い積層フィルムを得た。得られた積層フィルムの物性値を表１に示す。

【００２７】実施例３

実施例２で用いた低分子量ポリエチレン水性分散体（ＰＯ－２）に代えて、低分子量ポリエチレン水性分散体（ＰＯ－２）に更に１，４－ブタンジオール・コハク酸エステルの水性分散体（昭和高分子社製　商品名ビオノーレエマルジョンＯＬＸ－７５２７、融点：９３～９５℃、ガラス転移点：－４５℃、結晶化度：２０～３５%、比重：１．３、水生分散体の物性：固形分濃度：５０～５６重量%、ｐＨ：４～６、粘度：１００ＣＰ、粒径：１～２μｍ）を固形分中に１２重量%配合し、更に１００００ｒｐｍで１分間攪拌して得た水性分散体を用いる以外は実施例２と同様に行い積層フィルムを得た。得られた積層フィルムの物性値を表１に示す。

【００２８】比較例１

前記フィルム両面にコロナ放電処理した厚さ２５μｍの生分解性ポリエステル二軸延伸フィルムの物性値を比較例１として表１に示す。

【００２９】

【表１】

		実施例1	実施例2	実施例3	比較例1
低分子量ポリエチレン層		PO-1	PO-2	PO-2	なし
物性値					
透湿度	g/m^2・day	19	19	31	70
ヘイズ	%	61	88	92	4
光線透過率	%	36	11	7	69

【００３０】表１に示した結果から明らかなように、低分子量ポリエチレン層が積層されてなる生分解性ポリエステル二軸延伸積層フィルム（実施例１～実施例３）が、低分子量ポリエチレン層を有しない生分解性ポリエステル二軸延伸フィルムに比べ、透湿度が改良されている。

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(54) BIODEGRADABLE RESIN COMPOSITION

(57)Abstract:
PROBLEM TO BE SOLVED: To provide a biodegradable resin composition having an excellent disintegrable property in water and an excellent biodegradable property.
SOLUTION: This biodegradable resin composition comprising a biodegradable resin and polyethylene glycol in a total amount of 25 to 85 wt.% and an inorganic filler in an amount of 75 to 15 wt.% is characterized in that the polyethylene glycol having a number-average molecular weight of 5,000 to 30,000 is contained in an amount of 10 to 100 pts.wt. per 100 pts.wt. of the biodegradable resin.

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(54)【発明の名称】　生分解性樹脂組成物

(57)【要約】
【課題】　優れた水崩壊性、及び生分解性を有する生分解性樹脂組成物を提供する。
【解決手段】　生分解性樹脂とポリエチレングリコールの合計量２５～８５重量%、及び無機充填剤７５～１５重量%を含む生分解性樹脂組成物であって、前記生分解性樹脂１００重量部に対して、数平均分子量が５０００～３００００であるポリエチレングリコール１０～１００重量部を含むことを特徴とする生分解性樹脂組成物。

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【特許請求の範囲】

【請求項１】　生分解性樹脂とポリエチレングリコールの合計量２５～８５重量%、及び無機充填剤７５～１５重量%を含む生分解性樹脂組成物であって、前記生分解性樹脂１００重量部に対して、数平均分子量が５０００～３００００であるポリエチレングリコール１０～１１０重量部を含むことを特徴とする生分解性樹脂組成物。

【請求項２】　生分解性樹脂が、乳酸系ポリマー、及びポリアルキレンサクシネートから選ばれた少なくとも一種の樹脂であることを特徴とする請求項１記載の生分解性樹脂組成物。

【請求項３】　乳酸系ポリマーがポリ乳酸であることを特徴とする請求項２記載の生分解性樹脂組成物。

【請求項４】　ポリアルキレンサクシネートが、ポリブチレンサクシネート、ポリブチレンサクシネート・アジペート、ポリブチレンサクシネート・テレフタレート、及びポリエチレンサクシネートから選ばれた少なくとも一種の樹脂であることを特徴とする請求項２記載の生分解性樹脂組成物。

【請求項５】　無機充填剤が、炭酸カルシウム、硫酸バリウム、タルク、クレー、シリカ、及び雲母から選ばれた少なくとも一種の化合物であることを特徴とする請求項１記載の生分解性樹脂組成物。

【請求項６】　生分解性樹脂組成物が、請求項１記載の生分解性樹脂組成物を成形加工したシート、フィルム、及び容器から選ばれた少なくとも一種の成形物であることを特徴とする生分解性樹脂組成物。

【請求項７】　成形物が、生理用タンポンのアプリケータ、生理用ナプキン包装用フィルム、使い捨ておむつ用バックシート、及び医療用衣類から選ばれた少なくとも一種の衛生用品であることを特徴とする請求項６記載の樹脂組成物。

【請求項８】　２３℃の水に２日間浸漬したときの重量減少率が少なくとも１０%であり、且つ２３℃の水に１０分間浸漬したときの重量減少率が１０%未満であることを特徴とする請求項１～７のいずれか１項に記載の生分解性樹脂組成物。

【発明の詳細な説明】

【０００１】

【発明の属する技術分野】本発明は、生分解性樹脂組成物に関する。詳しくは、生理用タンポン、ナプキン、紙おむつ、及び医療用衣類等の衛生用品の成形用材料に適し、使用時は体液等により崩壊することがなく通常と全く同様に使用することができ、使用後トイレ、台所流し等に放流して水と接触した後には、崩壊及び分解する特性を有する生分解性樹脂組成物に関する。

【０００２】

【従来の技術】従来、生理用タンポン、ナプキン、使い捨ておむつ等は、トイレ等から放流処理することを想定して設計されていない。本来であれば、これらの物品、特に生理用品はトイレ内で装着、脱着がされることが多く、使用後はトイレットペーパーなどと同様に放流処理したほうが、簡単で汚れず形跡も残らないため好ましい。

【０００３】しかしながら、これらの物品の大部分は、一般的にポリエチレン、ポリプロピレン等の非分解性樹脂からなっており、放流することが出来ないため、使用者は別途設けられた箱などへ入れるか、持ち帰って別途処理する煩わしさがある。また、その後、ゴミとしての処理する際に、飛散したり、埋め立て処理される場合に分解しない等の問題もある。さらに、万が一トイレにて放流された場合は、下水処理系に蓄積されたり閉塞を引き起こす。また、分解されないためしばしば河川、海洋へ放出され環境への悪影響が大きくなる。このように、使用者にとっては処理の煩わしさ、処理時に手が汚れる等の不衛生さ、処理の形跡が残る等の心理的圧迫からの開放が望まれている。また、処理面からは、衛生面、環境への負加を改善する必要性が唱えられている。

【０００４】これらの問題を解決する方法として、特開平１０－１３１０８３号公報等には、水解紙が開示されている。しかし、これらは、紙を原料として用いており、衛生用品等のように風合いが重視される用途には不向きであった。紙ではなくプラスチックを用いた例として、特開昭６３－３０２８４５号公報、特開平５－２９５２１１号公報、特開平６－１３４９１０号公報、特公平７－５７２３０号公報などに、水溶性樹脂を用いた衛生用品の例示がある。これらの開示によれば、ポリビニルアルコールやポリ－３－ヒドロキシ酪酸等の水溶性樹脂を用いる事により、上記問題の解決を図っている。

【０００５】また、特開平６－２９９０７７号公報には、生分解性プラスチックス２０～８０重量%と水溶性熱可塑性樹脂８０～２０重量%を混合して得られる生分解性樹脂組成物の開示されている。しかしながら、これらの水溶性樹脂や生分解性樹脂は一般的に高価であり、使用量、添加量を増やすと価格が高くなり、衛生用品のような使い捨て用途には好ましくない。逆に、安価にするため添加量を減らすと、水崩壊性、生分解性が不足し、放流処理が出来ず好ましくない。さらには、本発明者らの知見では、これらの水溶性樹脂を含む樹脂組成物は経時的に水分を吸収しやすく、使用前に湿潤して表面がべとべとになったり、カビが発生したりするため、実使用上問題がある場合がある。

【０００６】

【発明が解決しようとする課題】本発明者らは、上記問題点を解決する手段として、特願２００１－１１４７７２号に係わる特許出願において、生分解性樹脂２５～８０重量%、無機充填剤７５～２０重量%、及び、前記生分解性樹脂１００重量部に対して水溶性樹脂３～１００重量部を含むことを特徴とする生分解性樹脂組成物を提案した。該樹脂組成物は、良好な水崩壊性、生分解性を

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有し、使用時には経時的な表面状態の悪化がない成形物を与える。しかしながら、該組成物を実際の使用形態であるタンポン用アプリケータなどに成形して評価した場合、水への溶解速度が比較的速いため、使用時に体内に成分が若干残存する可能性があり、また、成形物の保管中、経時的に硬化が進行することがあり、用途によっては必ずしも満足できない点があることが判った。

【０００７】本発明の目的は、上記の問題点に鑑み、優れた水崩壊性、及び生分解性を有しながらも、実使用時には成分が溶け出すことがほとんどなく経時的にも安定で、使用後、トイレ、台所流し等へ放流処理するだけで簡単に崩壊する生分解性樹脂組成物を提供することにある。

【０００８】

【課題を解決するための手段】本発明者らは、上記課題を達成するため鋭意検討した結果、特定量の生分解性樹脂、特定量の無機充填剤、及び特定量のポリエチレングリコールを含む生分解性樹脂組成物が、良好な水崩壊性、生分解性を有し、使用時には成分の溶けだしがほとんどなく、経時的にも安定な成形物を与えることを見出し、本発明を完成するに至った。

【０００９】すなわち、本発明は、生分解性樹脂とポリエチレングリコールの合計量２５～８５重量%、及び無機充填剤７５～１５重量%を含む生分解性樹脂組成物であって、前記生分解性樹脂１００重量部に対して、数平均分子量が５０００～３００００であるポリエチレングリコール１０～１１０重量部を含むことを特徴とする生分解性樹脂組成物である。

【００１０】本発明に係わる生分解性樹脂組成物の好ましい態様として、上記生分解性樹脂が、乳酸系ポリマー、及びポリアルキレンサクシネートから選ばれた少なくとも一種の樹脂である前記樹脂組生物が挙げられる。それらの内、乳酸系ポリマーとしてはポリ乳酸、ポリアルキレンサクシネートとしては、ポリブチレンサクシネート、ポリブチレンサクシネート・アジペート、ポリブチレンサクシネート・テレフタレート、及びポリエチレンサクシネートから選ばれた少なくとも一種の樹脂が挙げられる。また、上記無機充填剤として、炭酸カルシウム、硫酸バリウム、タルク、クレー、シリカ、及び雲母から選ばれた少なくとも一種の化合物が挙げられる。

【００１１】本発明に係わる前記各種の生分解性樹脂組成物は、これらを成形加工することにより、シート、フィルム、容器、又はこれらの組み合わせ成形物となすことができる。具体的成形物として、生理用タンポンのアプリケータ、生理用ナプキン包装用フィルム、使い捨ておむつ用バックシート、及び医療用衣類から選ばれた少なくとも一種の成形物が挙げられる。これらは、トイレ又は台所流しへ放流処理可能な衛生用品である。

【００１２】本発明の生分解性樹脂組成物の主な特性としては、２３℃の水に２日間浸漬したときの重量減少率が少なくとも１０%であり、且つ２３℃の水に１０分間浸漬したときの重量減少率が１０%未満であることが挙げられる。

【００１３】本発明の生分解性樹脂組成物は、特定量の生分解性樹脂、ポリエチレングリコール、及び無機充填剤を含むために、優れた水崩壊性、生分解性を有し、経時的にも表面状態の悪化がなく、しかも廉価である。また、使用時に成分が溶け出すことがなく、経時的にも安定である。そのため、上記衛生用品などの成形物として用いた場合、使用後の処理が非常に簡便で衛生的となり、処理後も環境への負加を減らすことが可能である。

【００１４】

【発明の実施の形態】以下、本発明について詳しく説明する。本発明の生分解性樹脂組成物は、特定量の生分解性樹脂、無機充填剤、及びポリエチレングリコールを混合することにより製造される。上記材料を混合した後、連続溶融混練を行うことが好ましい。

【００１５】本発明に用いる生分解性樹脂としては、乳酸系ポリマー、ポリブチレンサクシネート、ポリブチレンサクシネート・アジペート、ポリブチレンサクシネート・テレフタレート、ポリエチレンサクシネート、及びポリブチレンサクシーネート・カーボネート等のポリアルキレンサクシネート、ポリグリコール酸、ポリカプロラクトン、ポリヒドロキシ酪酸、ポリヒドロキシ吉草酸、ヒドロキシ酪酸・ヒドロキシ吉草酸共重合体等が挙げられる。これらの内、乳酸系ポリマー、ポリブチレンサクシネート、ポリブチレンサクシネート・アジペート、ポリブチレンサクシネート・テレフタレート、及びポリエチレンサクシネートが好ましい。

【００１６】上記乳酸系ポリマーとしては、ポリ乳酸、または乳酸と他のヒドロキシカルボン酸とのコポリマーが挙げられる。コモノマーとして用いられる他のヒドロキシカルボン酸として、グリコール酸、ヒドロキシ酪酸、ヒドロキシ吉草酸、ヒドロキシペンタン酸、ヒドロキシカプロン酸、ヒドロキシヘプタン酸等が例示される。

【００１７】好ましいポリ乳酸の分子構造は、Ｌ－乳酸またはＤ－乳酸いずれかの単位８５～１００モル%、さらに好ましくは該単位８５～９８モル%、及び、それぞれの対掌体の乳酸単位０～１５モル%、さらに好ましくは、該対掌体単位２～１５モル%を含むものである。また、乳酸と他のヒドロキシカルボン酸とのコポリマーは、Ｌ－乳酸またはＤ－乳酸いずれかの単位８５～１００モル%、好ましくは該単位８５～９８モル%、及び、他のヒドロキシカルボン酸単位０～１５モル%、好ましくは該単位２～１５モル%を含むものである。好ましい他のヒドロキシカルボン酸として、グリコール酸、ヒドロキシカプロン酸が挙げられる。

【００１８】これらの乳酸系ポリマーは、Ｌ－乳酸、Ｄ－乳酸および他のヒドロキシカルボン酸の中から必要と

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する構造のものを選んで原料とし、脱水重縮合することにより得ることができる。好ましくは、乳酸の環状二量体であるラクチド、グリコール酸の環状二量体であるグリコリド、およびカプロラクトン等から必要とする構造のものを選んで開環重合することにより得ることができる。

【００１９】ラクチドには、Ｌ－乳酸の環状二量体であるＬ－ラクチド、Ｄ－乳酸の環状二量体であるＤ－ラクチド、Ｄ－乳酸とＬ－乳酸とが環状二量化したメソ－ラクチド、およびＤ－ラクチドとＬ－ラクチドとのラセミ混合物であるＤＬ－ラクチドがある。本発明ではいずれのラクチドも用いることができる。ただし、主原料は、Ｄ－ラクチドまたはＬ－ラクチドが好ましい。

【００２０】本発明に用いられる好ましい乳酸系ポリマーは、上記組成のポリ乳酸、または乳酸と他のヒドロキシカルボン酸とのコポリマーであるが、それらは、次の（１）～（６）の方法により得ることができる。

（１）Ｌ－ラクチドをおよそ８５モル％以上とＤ－ラクチドおよび、またはグリコリドをおよそ１５モル％以下とを共重合させる、（２）Ｄ－ラクチドをおよそ８５モル％以上とＬ－ラクチドおよび、またはグリコリドをおよそ１５モル％以下とを共重合させる、（３）Ｌ－ラクチドをおよそ７０モル％以上とＤＬ－ラクチドおよび、またはグリコリドをおよそ３０モル％以下とを共重合させる、（４）Ｌ－ラクチドをおよそ７０モル％以上とメソ－ラクチドおよび、またはグリコリドをおよそ３０モル％以下とを共重合させる、（５）Ｄ－ラクチドをおよそ７０モル％以上とＤＬ－ラクチドおよび、またはグリコリドをおよそ３０モル％以下とを共重合させる、

（６）Ｄ－ラクチドをおよそ７０モル％以上とメソ－ラクチドおよび、またはグリコリドをおよそ３０モル％以下とを共重合させる。

【００２１】これらの乳酸系ポリマーは高分子量であることが好ましく、濃度０．５ｇ／ｄｌのクロロホルム溶液（２５℃）の固有溶液粘度が１～１０であることが好ましく、さらに好ましくは３～７である。固有溶液粘度が１未満であると、溶融粘度が低すぎて押出機のダイのスリットから流下し、成形し難いばかりでなく、脆すぎて取扱が困難であり、また、１０を超えると溶融粘度が高過ぎて押出成形性等が悪くなるので好ましくない。

【００２２】ラクチド、または、ラクチドとグリコリドを重合させて、短時間で高分子量のポリマーを得るためには触媒を用いるのが好ましい。このような重合触媒としては、この重合反応に触媒効果を示す各種のものが使用できる。例えば、公知なものとして、オクタン酸第一錫、四塩化錫、塩化亜鉛、四塩化チタン、塩化鉄、三フッ化ホウ素エーテル錯体、塩化アルミニウム、三フッ化アンチモン、酸化鉛等の主として多価金属を含む化合物が挙げられ、中でも錫化合物または亜鉛化合物が好ましく使用される。錫化合物の中ではオクタン酸第一錫が特に好ましい。使用量はラクチド、または、ラクチドとグリコリドの総量に対して、およそ０．００１～０．１重量％程度が好ましい。

【００２３】また、重合の際には、公知の連鎖増大剤を用いることができる。連鎖増大剤としては、ラウリルアルコール等の高級アルコール類、乳酸やグリコール酸等のヒドロキシ酸類が好ましく用いられる。連鎖増大剤の共存により、重合速度が大きくなるので短時間でポリマーを得ることができる。また、連鎖増大剤の量を加減することによりポリマーの分子量を調節することもできる。しかし、連鎖増大剤の量を多くしすぎると生成ポリマーの分子量は小さくなる傾向があるので、連鎖増大剤を使用する場合には、その量はラクチド、または、ラクチドとグリコリドの総量に対して０．１重量％以下であることが好ましい。

【００２４】重合または共重合するに当たって、溶媒を用いても用いなくてもよいが、高分子量のポリマーを得るには、ラクチドまたはグリコリドを溶融した状態で塊状重合することが好ましい。重合温度は、溶融重合の場合には原則的にはモノマーであるラクチド、またはラクチドとグリコリドの融点（９０℃付近）以上の温度であればよい。また、例えば、クロロホルム等の溶媒を用いる溶液重合の場合には、ラクチド、または、ラクチドとグリコリドの融点以下の温度で重合することが可能である。いずれの場合も、２５０℃を超えると生成ポリマーの分解が起こるので好ましくない。ポリ乳酸の代表的な市販品として、三井化学（株）製、商品名：レイシアが挙げられる。

【００２５】ポリアルキレンサクシネートとしては、ポリブチレンサクシネート、ポリブチレンサクシネート・アジペート、ポリブチレンサクシネート・テレフタレート、ポリエチレンサクシネート、及びポリブチレンサクシーネート・カーボネートが好ましい。ポリブチレンサクシネートの市販品として、昭和高分子（株）製、商品名：ビオノーレ１００１、同ビオノーレ１０２０、同ビオノーレ１９０３等が挙げられる。また、ポリブチレンサクシネート・アジペートの市販品として、昭和高分子（株）製、商品名：ビオノーレ３００１、同ビオノーレ３０２０等が挙げられる。

【００２６】ポリブチレンサクシネート・テレフタレートの市販品として、ＢＡＳＦ社製、商品名：エコフレックス、ＤｕＰｏｎｔ社製、商品名：バイオマックス等が挙げられる。ポリエチレンサクシネートの市販品として、（株）日本触媒製、商品名：ルナーレＳＥが挙げられる。ポリブチレンサクシネート・カーボネートの市販品として、三菱ガス化学（株）製、商品名：ユーペックが挙げられる。

【００２７】本発明の生分解性樹脂組成物は、上記生分解性樹脂１００重量部に対し可塑剤を１～５０重量部含んでもよい。可塑剤として、例えば、ジーｎーオクチル

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フタレート、ジ－２－エチルヘキシルフタレート、ジベンジルフタレート等のフタル酸誘導体、ジイソオクチルフタレート等のイソフタル酸誘導体、ジ－ｎ－ブチルアジペート、ジオクチルアジペート等のアジピン酸誘導体、ジ－ｎ－ブチルマレート等のマレイン酸誘導体、トリ－ｎ－ブチルシトレート等のクエン酸誘導体、モノブチルイタコネート等のイタコン酸誘導体、ブチルオレート等のオレイン酸誘導体、グリセリンモノリシノレート等のリシノール酸誘導体、トリクレジルフォスフェート、トリキシレニルフォスフェート等のリン酸エステル系可塑剤、アセチルクエン酸トリエチル、アセチルクエン酸トリブチル、乳酸、直鎖状乳酸オリゴマー、環状乳酸オリゴマーまたはラクチド等が例示できる。特に分子内に２個以上のカルボン酸エステル基を有する、クエン酸エステル、グリセリンエステル、フタル酸エステル、アジピン酸エステル、セバシン酸エステル、アゼライン酸エステルおよびトリエチレングリコールエステルから選ばれた少なくとも１種のエステル化合物であることが好ましい。これらの可塑剤は単独でも良いし、また２種以上を混合して用いてもよい。

【００２８】かかる可塑剤を乳酸系ポリマーに添加することにより、乳酸系ポリマーは効果的に可塑化され、得られる樹脂組成物は柔軟性を帯びる。樹脂組成物中の可塑剤の量が１重量%以上になると柔軟性がはっきり現れるようになり、５０重量%を超えると、該組成物を溶融押出する時の成形性が悪くなり、また、得られた成形物の強度が弱くなったり、経時的に可塑剤が浮き出してくるため好ましくない。

【００２９】本発明の生分解性樹脂組成物には、上記生分解樹脂と、特定量の無機充填剤、及び生分解性樹脂に対し特定量のポリエチレングリコールが添加される。ポリエチレングリコールとしては、ポリエチレングリコール、またはその変性物が挙げられる。ポリエチレングリコールの数平均分子量は、本発明の樹脂組成物を成形した際の、水への溶解速度、及び経時的な安定性に影響を及ぼす。数平均分子量が低すぎると、水への溶解速度が速すぎ、例えばタンポンアプリケータとして用いた場合、使用時に体内に成分が残存したり、極端な場合、体内で崩壊が進行する可能性が大きくなる。また、成形後に保管する際、経時的に樹脂が硬化するなどの問題が生じる。

【００３０】かかる点を考慮すると、ポリエチレングリコールの数平均分子量は５０００以上であることが好ましい。更に好ましくは７０００以上、特に好ましくは９０００以上である。数平均分子量の上限については、高ければ高いほど好ましい。原料の入手の容易なことを考慮すると、通常、数平均分子量の上限は３００００程度である。好ましくは２００００程度である。上記ポリエチレングリコールは数平均分子量の異なるものを混合しても良い。混合前の個別の数平均分子量が５０００以下であっても、混合後の全体としての数平均分子量（以下、Ｍｎという）が５０００以上であれば問題はない。

【００３１】ポリエチレングリコールの市販品として、三洋化成工業（株）製、商品名：ＰＥＧ－１０００（Ｍｎ：１０００）、同－４０００Ｓ（Ｍｎ：３３００）、同－６０００Ｓ（Ｍｎ：８３００）、同－１００００（Ｍｎ：１１０００）、同－１３０００（Ｍｎ：１３０００）、同－２００００（Ｍｎ：２００００）等が挙げられる。ポリエチレングリコールのＭｎの算出方法としては、まず無水酢酸・ピリジン法により水酸基価を求め、その水酸基価からＭｎを算出する。詳細については後述の実施例に記載する。

【００３２】本発明の生分解性樹脂組成物は、上記生分解性樹脂に対し、特定量の上記ポリエチレングリコールを含む。ポリエチレングリコールの含有量は、樹脂組成物の水崩壊性、生分解性に影響を及ぼす。水溶性樹脂の含有量が少な過ぎると、水崩壊性が不足し、その結果、生分解性も抑制されることとなり好ましくない。逆に、水溶性樹脂の含有量が多過ぎると、水崩壊性は良好であるが、樹脂組成物の水親和性が過剰となり、水と隔離された状態であっても、空気中の湿気により経時的に成形物の表面がべとべとしたり、カビが発生したりすることがある。また、樹脂組成物が高価となるため、廉価性が必要とされる衛生用品のような使い捨て用途には好ましくない。さらに、結果として相対的に生分解性樹脂の量が低下するため、強度が低下する問題がある。かかる点を考慮すると、生分解性樹脂１００重量部に対して、ポリエチレングリコール１０～１１０重量部を含むことが好ましい。より好ましい添加量としては１５～１０５重量部である。

【００３３】本発明に用いられる無機充填剤としては、炭酸カルシウム、炭酸マグネシウム、炭酸バリウム、硫酸マグネシウム、硫酸バリウム、硫酸カルシウム、酸化亜鉛、酸化マグネシウム、酸化カルシウム、酸化チタン、酸化マグネシウム、アルミナ、水酸化アルミニウム、ヒドロキシアパタイト、シリカ、マイカ、タルク、カオリン、クレー、ガラス粉、アスベスト粉、ゼオライト、珪酸白土、雲母等が挙げられる。これらの内、炭酸カルシウム、酸化マグネシウム、硫酸バリウム、シリカ、珪酸白土等が好ましく用いられる。より好ましくは、炭酸カルシウム、硫酸バリウムである。これらの無機充填剤は単独で用いても良いし、２種以上混合して用いても良い。

【００３４】上記無機充填剤と生分解性樹脂との配合比は、得られる樹脂組成物の分解性、表面状態、廉価性に影響を及ぼす。無機充填剤の含有量が多過ぎると、樹脂成分が極端に低下し強度が不足するため好ましくない。また、樹脂への混練も困難となり生産性が低下する。逆に少な過ぎると、強度や混練の難易性には問題がないが、水崩壊性が低下する上に、樹脂組成物の廉価性が損

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なわれるために好ましくない。また、無機充填剤は多少なりとも吸着性があり、樹脂組成物が空気中の湿気により経時的に表面がべとべとしたり、カビが発生したりするのを防ぐ効果があるが、その効果が低下するために好ましくない。かかる観点から、本発明の生分解性樹脂組成物は、生分解性樹脂とポリエチレングリコールの合計量２５～８５重量%、及び無機充填剤７５～１５重量%を含むことが好ましい。より好ましくは、生分解性樹脂とポリエチレングリコールの合計量３０～９０重量%、無機充填剤７０～１０重量%である。

【００３５】これらの無機充填剤の平均粒径としては、０．３～１０μmのものが好ましい。さらに好ましくは０．５～５μmである。平均粒径が１０μmを超えると樹脂組成物の表面状態が荒くなる上に、美観も損ねるため好ましくない。また、フィルムやシートなどへの成形の際、穴があくなどの不良が発生しやすく生産性、作業性等の点でも好ましくない。平均粒径が０．３μm未満の場合、無機充填剤の凝集が発生しやすく、均一な練り込みが出来ないため好ましくない。また、無機充填剤の嵩密度が低くなり、混練時の作業性が低下し生産性が低下するため好ましくない。

【００３６】無機充填剤は、樹脂との分散性を向上させるために表面処理を行っても良い。但し、表面処理により無機充填剤の吸湿性が損なわれ、樹脂組成物の経時的な表面状態の悪化を抑制できない場合はこの限りではない。表面処理剤としては、例えば、ステアリン酸、ラウリン酸等の高級脂肪酸またはそれらの金属塩等を挙げることができる。本発明の生分解性樹脂組成物には、その他に本発明の目的を妨げない範囲で分散剤、顔料、安定剤、酸化防止剤、着色剤、紫外線吸収剤等の他の添加剤を添加してもよい。

【００３７】上記の通り、本発明の生分解性樹脂組成物は、特定量の生分解性樹脂、ポリエチレングリコール、及び無機充填剤を含み、優れた水崩壊性、生分解性樹脂を有し、経時的にも表面状態の悪化がなく、しかも廉価である。優れた水崩壊性及び生分解性を有する理由は定かではないが、一応、次ぎのような機構を推定する。

【００３８】本発明の生分解性樹脂組成物が水中あるいは土壌中に廃棄された場合、先ず、樹脂組成物中のポリエチレングリコールが水に溶解する。その結果、樹脂組成物中には空隙が多くでき、ポリエチレングリコールの溶解が進むにつれて破断され、場合によっては粒状にまで崩壊する。本発明の生分解性樹脂組成物には無機充填剤が多く含まれているため、この崩壊の際に同時に無機充填剤が脱落することにより、ますます崩壊が促進される。その結果、樹脂組成物中には空隙が多く形成され、樹脂の表面積が増えることとなり、残った生分解性樹脂は比較的速やかに分解するものと推定される。

【００３９】次いで、本発明の生分解性樹脂組成物の製造方法を例示する。例えば、生分解性樹脂、無機充填剤、ポリエチレングリコール、必要に応じて可塑剤、分散剤、安定剤などその他の添加物をヘンシェルミキサー、スーパーミキサー、タンブラー型ミキサー等を用いて混合した後、一軸または二軸スクリュー型押出機を用いて連続混練する。ここで、生分解性樹脂中への無機充填剤、ポリエチレングリコールの分散性をより向上させるためには、二軸押出機の方が好ましい。

【００４０】押出温度は、好ましくは１００～２７０℃の範囲、更に好ましくは１３０～２５０℃の範囲である。１００℃未満では、押出安定性が得難く、また過負荷に陥りやすく、２７０℃を超えると、生分解性樹脂やポリエチレングリコールの分解が激しくなるので好ましくない。

【００４１】本発明の生分解性樹脂組成物から成形し得る成形体としては、例えば、生理用タンポン用アプリケータ、生理用ナプキン包装用フィルム、生理用ナプキン用バックシート、使い捨てオムツ用バックシート、使い捨てオムツ包装用フィルム等の衛生用品の容器、フィルム、シートが挙げられる。例えば、生理用タンポンアプリケータは、直径が２～３０mm、長さが１０～２００mm、厚みが０．０５～５mmの筒状容器であって、射出成形で成形したり、シート状物を得た後、筒状に丸める等により成形できる。

【００４２】また、フィルム、シートを得る場合には、Ｔダイなどが装着された押出機により溶融押出成形することにより、厚み０．０１～５mmに成形可能である。これらの成形物は、その分解速度を高めるために薄く成形することが好ましいが、強度等を満足させるように自由に調整可能である。好ましい厚みは、タンポン用アプリケータとしては０．１～５mm厚、より好ましくは０．２～２mm厚である。また、フィルムとしては、０．０１～０．１mm厚、より好ましくは０．０２～０．０７mm厚である。

【００４３】上記製造方法により得られる本発明の生分解性樹脂組成物、及びその成形物は、２３℃の水に２日間浸漬したときの重量減少率が少なくとも１０%であり、且つ２３℃の水に１０分間浸漬したときの重量減少率が１０%未満である。該２日間浸漬したときの重量減少率の上限は特に制限されるものではないが、本発明の範囲では主に生分解性樹脂や無機充填剤の配合比に影響され、約９０%程度である。該２日間浸漬したときの重量減少率が１０%未満であるような従来の樹脂組成物は、水崩壊性が不十分であり、上記条件を経過した後の崩壊、分解が極めて長期間に及ぶため好ましくない。より好ましくは、２日間浸漬したときの重量減少率が少なくとも１２%である。

【００４４】さらに、該２３℃の水に１０分間浸漬したときの重量減少率は１０%未満である。１０分間浸漬したときの重量減少率が１０%以上の場合、水への溶解速度は速すぎるため、例えばタンポンアプリケータなどに

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用いた場合、体内に成分が残存したり、極端な場合体内で崩壊が進む可能性が高くなり好ましくない。より好ましくは、１０分間浸漬したときの重量減少率は８％未満である。

【００４５】該２日間浸漬したときの重量減少率、及び１０分間浸漬したときの重量減少率は、ポリエチレングリコールの数平均分子量、及びその含有率やその他の添加剤も含んだ含有率と大きな相関がある。これらの３種の材料を本発明で規定する範囲で含むことにより、優れた水崩壊性、及び生分解性樹脂が発現するのである。その上、経時的にも表面状態の悪化及び樹脂の硬化などの現象がなく、実使用時には成分が溶け出すことがほとんどないため、使い捨ての衛生材料として極めて有用である。

【００４６】

【実施例】以下、実施例を示して本発明についてさらに詳細に説明する。なお、この実施例で用いた評価方法は、以下の通りである。

【００４７】（１）表面状態

実施例で得られた樹脂シートから、縦横それぞれ１０ｃｍ、厚み０．５ｍｍの試料を調製する。試料を４０℃、相対湿度６０％の恒温恒湿槽に２４時間放置した後の表面状態を観察する。評価基準は以下の通りである。○：表面状態の変化はほとんど認められない。ヌメリ感なし。△：表面状態の変化が若干認められる。ヌメリ感がある。×：表面がべとついている。

【００４８】（２）２日間重量減少率（％）

前項（１）と同様の試料を室温において、２３℃の純水５リットル中に２日間浸漬した後に取り出す。取り出した後、水道流水にて表面のヌメリ等がなくなるまで洗う。洗浄後、室温で２時間傾斜放置し水切りを行う。その後、シリカゲルを入れたデシケータ中で１２時間乾燥させる。下記数式（１）により重量減少率を算出する。１０枚の試料について測定し、その平均値を算出する。

$W_R = [(W_1 - W_2) / W_1] \times 100$ ・・（１）

ここで、W_R：重量減少率（％）、W_1：浸漬前の重量、W_2：浸漬後の重量。

【００４９】（３）１０分間重量減少率（％）

前項（１）と同様の試料を室温において、２３℃の純水５リットル中に１０分間浸漬した後に取り出す。取り出した後、室温で２時間傾斜放置し水切りを行う。その後、シリカゲルを入れたデシケータ中で１２時間乾燥させる。重量減少率を前項（２）と同様の方法で算出する。

【００５０】（４）無機充填剤の平均粒径（μｍ）

レーザー回折散乱法粒度分布測定器（日機装（株）製、形式：マイクロトラックＸ－１００）を用いて計測し、平均粒径（μｍ）を測定する。

【００５１】（５）経時強度

実施例で得られた試料（厚み０．５ｍｍのシート）を５０℃のオーブンに４日間放置したのち、１８０°に折り曲げ、下記の基準に従って評価を行う。

○：折り曲げ箇所が破壊せず、亀裂が生じない。△：折り曲げ箇所に亀裂が生じる。×：２つ以上の断片に破壊する。

【００５２】（６）ポリエチレングリコールの数平均分子量

水酸基価を測定し、得られた水酸基価から数平均分子量を算出する。

＜水酸基価（ＫＯＨｍｇ／ｇ）の測定＞［表１］に基づき、試料を三角フラスコに精秤し、無水酢酸・ピリジン混合試薬（無水酢酸１容にピリジン４容の割合で混合したもの）５ｍｌをホールピペットにて加える。９５～１００℃の油浴に上記三角フラスコを浸け、冷却器を付け１時間反応する。イオン交換水１ｍｌを加え、よく攪拌し、さらに１０分間油浴上で放置する。油浴から取り出し、さらに１０分間放置する。０．５％フェノールフタレインを指示薬として、０．５ｍｏｌ／Ｌ水酸化カリウム標準溶液で滴定する。以上の操作を２回行うと同時に、併行して空試験も行う。以下の式に従い水酸基価を求め、２回の平均をとる。

$OH = [\{ (B - A) \times 28.05 \times f \} / S]$

ここで、ＯＨ：水酸基価（ＫＯＨｍｇ／ｇ）、Ｂ：空試験に要した０．５ｍｏｌ／Ｌ水酸化カリウム標準溶液のｍｌ数、Ａ：本試験に要した０．５ｍｏｌ／Ｌ水酸化カリウム標準溶液のｍｌ数、ｆ：０．５ｍｏｌ／Ｌ水酸化カリウム標準溶液の力価、Ｓ：試料採取量（ｇ）。

【００５３】尚、試料が遊離酸を含むときは、ＪＩＳＫ－１５５７－１９７０に規定される方法により酸価（ＫＯＨｍｇ／ｇ）を測定して、次の式により修正する。

修正ＯＨ＝ＯＨ＋酸価

ここで、修正ＯＨ：修正水酸基価（ＫＯＨｍｇ／ｇ）、ＯＨ：滴定で得られた水酸基価（ＫＯＨｍｇ／ｇ）。

【００５４】

【表１】

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水酸基価（KOHmg／g）	試料採取量（g）
５０未満	３．０～５．０
５０以上１００未満	２．０～３．０
１００以上１５０未満	１．５～２．０
１５０以上２００未満	１．０～１．５
２００以上２５０未満	０．８～１．０
２５０以上３００未満	０．６～０．８
３００以上４００未満	０．５～０．６

【００５５】＜数平均分子量の算出＞下記数式により算出する。

Ｍｎ＝（５６１００×２）／ＯＨ

ここで、Ｍｎ：数平均分子量、ＯＨ：滴定で得られた水酸基価又は上記修正水酸基価（ＫＯＨｍｇ／ｇ）。

調製例１

＜ジグリセリンテトラアセテートの調製＞ジグリセリン８４ｇ、及び無水酢酸４１５ｇを反応フラスコに入れ、強酸性イオン交換樹脂（ダウケミカル社製、商品名：ダウエックスＭＳＣ－１）２．５ｇを追加し、撹拌しながら８０～９０℃で１時間反応を行った。冷却後、イオン交換樹脂を濾別し、生成した酢酸および未反応の無水酢酸を減圧留去し、ジグリセリンテトラアセテート１６１．５ｇを得た。これは、一般式（１）におけるR_1～R_4が全て炭素数２のアシル基、ｎは１である、酸価が０．２、水酸基価が３．１である。

【００５６】実施例１～１６、比較例１～８

生分解性樹脂、可塑剤、ポリエチレングリコール、無機充填剤を〔表２〕～［表４］に示す銘柄、割合にて混合し、分散剤としてステアリン酸カルシウム（日東化成（株）製、商品名：Ｃａ－Ｓｔ）を上記樹脂組成物１００重量部に対して１重量部加え、ヘンシェルミキサーを用いて室温で混合して生分解性樹脂組成物を製造した。得られた樹脂組成物を１９０℃において、２軸スクリュー押出機にて混練した。さらに、混練物を１８０℃、１０ＭＰａにおいてプレス後、３０℃にてクエンチして、厚さ０．５ｍｍのシート状に加工した。得られたシートについて上記方法に従って物性を評価し、その結果を〔表２〕～［表４］に示す。

【００５７】

【表２】

		実施例１	実施例２	実施例３	実施例４	実施例５	実施例６	実施例７	実施例８
樹脂	種類	ＰＬＡ	ＰＬＡ	ＰＬＡ	ＰＬＡ	ＰＬＡ	ＰＬＡ	ＰＬＡ	ＰＬＡ
	量（重量部）	５０	５０	５０	５０	４４	４２	４２	４０
ＰＥＧ	種類	６０００Ｓ	１００００	１３０００	２００００	１００００	１００００	２００００	２００００
	Ｍｎ	８３００	１１０００	１３０００	２００００	１１０００	１１０００	２００００	２００００
	量（重量部）	１４	１４	１４	１４	１４	２２	２２	２２
	対樹脂100(重量部)	２８	２８	２８	２８	３２	５２	５２	５５
可塑剤	種類	－	－	－	－	－	－	－	－
	量（重量部）	－	－	－	－	－	－	－	－
無機充填剤	種類	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$BaSO_4$
	量（重量部）	３６	３６	３６	３６	４２	３６	３６	３８
２日間重量減少率	（重量%）	１３	１３	１２	１１	１３	２１	１８	１８
１０分間重量減少率	（重量%）	６	４	３	０	３	６	１	１
経時強度	－	△	○	○	○	○	○	○	○
表面状態	－	○	○	○	○	○	○	○	○

【００５８】

【表３】

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		実施例9	実施例10	実施例11	実施例12	実施例13	実施例14	実施例15	実施例16
樹脂	種類	PLA	PLA	PLA	PLA	PLA	PBS	PLA/m-PBT	PLA
	量(重量部)	40	70	30	47	50	50	25/25	40
PEG	種類	20000	20000	20000	10000	10000	10000	10000	20000
	Mn	20000	20000	20000	11000	11000	11000	11000	20000
	量(重量部)	35	13	14	20	14	14	14	40
	対樹脂100(重量部)	88	19	47	43	28	28	28	100
可塑剤	種類	−	−	−	A	−	−	−	−
	量(重量部)	−	−	−	3	−	−	−	−
無機充填剤	種類	$CaCO_3$	$CaCO_3$	$BaSO_4$	$CaCO_3$	talc	$CaCO_3$	$CaCO_3$	$CaCO_3$
	量(重量部)	25	17	56	30	36	36	36	20
2日間重量減少率	(重量%)	28	11	12	19	13	13	13	35
10分間重量減少率	(重量%)	2	1	1	5	3	4	4	4
経時強度	−	○	○	○	○	○	○	○	○
表面状態	−	○	○	○	○	○	○	○	○

【0059】

【表4】

		比較例1	比較例2	比較例3	比較例4	比較例5	比較例6	比較例7	比較例8
樹脂	種類	PLA	PLA	PLA	PLA	PLA	PLA	PLA	PLA
	量(重量部)	50	50	42	42	60	15	50	25
PEG	種類	1000	4000S	1000	4000S	10000	10000	10000	10000
	Mn	1000	3300	1000	3300	11000	11000	11000	11000
	量(重量部)	14	14	22	22	35	5	1	39
	対樹脂100(重量部)	28	28	52	52	58	33	2	156
可塑剤	種類	−	−	−	−	−	−	−	−
	量(重量部)	−	−	−	−	−	−	−	−
無機充填剤	種類	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$
	量(重量部)	36	36	36	36	5	80	49	36
2日間重量減少率	(重量%)	14	14	22	22	30	−	0	33
10分間重量減少率	(重量%)	11	8	18	12	8	−	0	10
経時強度		×	×	×	×	○	−	×	×
表面状態		×	△	×	△	×	−	○	×

【0060】<表2～表4の記載の説明>PLA：ポリ乳酸（三井化学（株）製、商品名：レイシア）、PBS：ポリブチレンサクシネート（昭和高分子（株）製、商品名：ビオノーレ1001）、m－PBT：ポリブチレンサクシネート・テレフタレート（BASF社製、商品名：エコフレックス）、A：可塑剤（調製例1で得られたジグリセリンテトラアセテート）、PEG：ポリエチレングリコール［三洋化成工業（株）製、商品名：PEG－1000（Mn；1000）、同－4000S（Mn；4000）、同－6000S（Mn；6000）、同－10000（Mn；10000）、同－13000（Mn；13000）、同－20000（Mn；20000）］、$CaCO_3$：重質炭酸カルシウム（同和カルフィン（株）製、商品名：SST－40、平均粒径2μm）、$BaSO_4$：沈降性硫酸バリウム（バライト工業（株）製、商品名：ST、平均粒径0．9μm）。

【0061】実施例17

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実施例１～１６で得られた、厚み０．５mmの各シートを筒状に丸めて、直径１０mm、長さ５０mmの筒状の生理用タンポンアプリケータを製造した。

【００６２】

【発明の効果】本発明により提供される生分解性樹脂組成物は、優れた水崩壊性及び生分解性を有し、経時的にも表面状態の悪化がなく、しかも廉価である。そのため、本発明の生分解性樹脂組成物からの成形物は、使用時は体液等により崩壊することがなく通常と全く同様に使用でき、使用した後トイレ、台所流し等に放流した場合、水崩壊及び生分解する効果を奏する。その結果、環境への負加を減らすことが可能である。従って、生理用タンポンのアプリケータ、生理用ナプキン包装用フィルム、使い捨て紙おむつのバックシート、及び医療用衣類等の衛生用品や、キッチンタオル、掃除用タオル、ワイピングクロス等の生活用品の分野において好適に使用される。

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(54) Title: BIODEGRADABLE POLYMER BLEND CONTAINING 3-HYDROXYBUTYRATE/3-HYDROXYVALERATE COPOLYMER

(57) Abstract

A biodegenerable plastics composition comprises at least one synthetic polymer selected from the class consisting of polyethylene, polypropylene, polystyrene and an ethylene copolymer, blended with 10 to 90 % w/w on said composition of a biodegradable material at least 20 % w/w of which is microbiologically produced copolymer containing 3-hydroxybutyrate and 3-hydroxyvalerate to the extent preferably of 3-25 mol %.

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BIODEGRADABLE POLYMER BLEND CONTAINING 3-HYDROXYBUTYRATE/3-HYDROXYVALERATE COPOLYMER

This invention relates to biodegenerable polymer blends comprising 3-hydroxybutyrate copolymer.

There is an increasing need for plastics materials which degenerate after use to environmentally acceptable waste products when in contact with microbiologically active systems such as soil, compost, activated sludge and sediment.

Conventional plastics materials containing synthetic polymers such as low density polyethylene (LDPE) and ethylene vinyl acetate (EVA) generally do not degenerate, i.e. disintegrate or degrade, through microbiological action in time periods of less than five years.

An LDPE-containing biodegenerable composite comprising LDPE and starch has been proposed. When in the form of a film, it is believed that the composite is degenerable by microbiological action within five years. Unfortunately, high levels of starch have been shown to be required before a suitable biodegenerable composite is obtained. The presence of high levels of starch have an unacceptable detrimental effect on the physical and processing characteristics of the composite. An ethylene-acrylic acid copolymer has been used to improve the physical properties of LDPE-starch composite, but only at low starch levels, at which levels the composite is not suitably biodegenerable.

We have found that a biodegenerable plastics composition having satisfactory physical properties comprises a microbiologically produced copolymer comprising 3-hydroxybutyrate and at least one of polyethylene, polypropylene, polystyrene, and an ethylene copolymer.

Accordingly to the invention a biodegenerable plastics composition comprises at least one synthetic polymer selected from the class consisting of polyethylene, polypropylene, polystyrene and ethylene copolymer blended with 10 to 90% w/w on said composition of a biodegradable material, at least 20 % w/w of which is microbiologically produced copolymer "PHBV" containing 3-hydroxybutyrate and 3-hydroxyvalerate.

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Preferably the biodegradable material represents between 20 and 50 % w/w of the composition and at least half thereof is said microbiologically produced copolymer. Compositions showing particularly good biodegenerable characteristics contain at least 20% w/w of said microbiologically produced copolymer.

The ethylene copolymer may be one or more copolymers of ethylene with one or more carboxylic derivatives of ethylene, especially acrylic acid, methacrylic acid, maleic anhydride, ethyl acrylate and vinyl acetate (EVA). A particularly preferred ethylene copolymer is EVA, especially containing 5 to 30 w/w % of vinyl acetate on the total of ethylene and vinyl acetate as illustrated in Examples 3 and 4 below. The EVA preferably has a melt index (ASTMD 1238) in the range 1.0 to 5.0, especially 1.5 to 3.5, g/10 min. It is advantageous for some uses to provide the EVA as a mixture with wax grade EVA in proportions in the range 90:10 to 40:60.

The composition , especially if containing high levels (at least 20% w/w) of microbiologically produced copolymer may also contain one or more biodegradable or environmentally acceptable improvers to optimise characteristics in respect of particular uses. Such improvers include plasticisers, processability improving agents and toughening agents. Examples of such plasticisers are glycerol triesters such as triacetin, glycerol tributyrate and glycerol tricaproate, in particular triacetin. An example of a biodegradable toughening agent is a crystalline polyester such as polycaprolactone.

The composition may also contain at least one biodegradable filler such as starch or cellulose, and/or environmentally acceptable filler such as calcium carbonate or talc.

The presence of at least one improver and/or filler is particularly preferred where the total biodegradable material represents at least 40 % w/w of the composition; and then it is particularly preferred that the microbiologically produced copolymer represent 35 to 65 % w/w of said biodegradable material.

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The physical properties of the composition may be altered by varying the relative proportions of the constituents. Thus, the Young's Modulus of such plastics compositions may vary between 70 and 225 (MPa), the Flexural Modulus may vary between 200 and 500 (Mpa), and the Extension at Break may vary between 50 and 500%. Particularly useful plastics compositions exhibit physical properties within at least two of the above ranges.

Particularly useful compositions comprising a binary blend of said biodegradable material with EVA contain between 5 and 50%, particularly between 10 and 40%, w/w of EVA.

A particularly useful composition comprises a ternary blend of (1) said biodegradable material, (2) at least one polymer selected from polyethylene, polypropylene and polystyrene, and (3) ethylene copolymer as hereinbefore defined. A particularly preferred such composition comprises said biodegradable material, polyethylene and EVA. The preferred such composition contains between 5 and 30 % w/w polyethylene (especially low density polyethylene) LDPE, and between 5 and 20 % w/w of EVA.

The microbiologically produced copolymers of 3-hydroxybutyrate and 3-hydroxyvalerate herein termed PHBV, may include minor proportions of other oxy-alkanoate residues. The PHBV copolymers show better compatibility with LDPE and EVA as compared with PHB homopolymer. Especially preferred are PHBV copolymers containing at least 2 especially at least 4, mol % of 3-hydroxyvalerate, and more particularly those copolymers containing 3 and 25, especially between 5 and 15, mol %.

The molecular weight of the PHBV in the composition is preferably at least 300000, especially at least 500000 and up to the limit of convenient processability, for example 1.5 million. Such molecular weights are characteristic of the polymer as now available commercially.

The composition may additionally contain one or more components such as fibrous fillers (which may be biodegenerable), pigments, anti-oxidants (other than those inhibiting biodegeneration), antistatic agents, stabilisers, foaming agents,

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slip agents, antiblocking agents and others commonly used in plastics processing. To facilitate processing and development of crystallinity of the PHBV, the composition may contain a nucleating agent as described below.

In all, the choice of constituents and their proportions is preferably such that articles made from the composition are denser than fresh water.

The invention provides also methods of making the composition by mixing the constituents in conditions in which at least one of its polymeric constituents is at least partly in liquid form. Examples of such methods are solution blending, slurry blending and melt blending. Preferably the constituents of the mixture are mechanically worked together, for example by screw-extrusion, roller-milling or kneading. The product of such mechanical working is typically formed into granules suitable for processing to shaped articles. The molecular weight of the microbiologically produced copolymer as introduced is preferably at least 500000, especially at least 800000, to allow for a decrease during melt processing. The mixture may contain processing aids such as one or more nucleating agents; and processing conditions are chosen preferably so as to maximise the development of crystallinity, to exploit this known property of the PHBV copolymer. Examples of nucleating agents are boron nitride, calcium carbonate and metal soaps such as zinc stearate.

In a further aspect the invention provides a process of making a shaped article by confining the composition to shaping surfaces. Methods of confining include casting from melt into a surface or in a mould, injection moulding , blow moulding and extrusion. A particularly useful method comprises extrusion of melt through a flat or circular die to give a flat or tubular film. The film may be subjected to stretching in the extrusion direction or transversely thereto or both, possibly with application of heat; for tubular film such stretchings may be effected at least partly by pneumatic pressure. As a sub-genus

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of extrusion through a circular due to give tubular film, the method may employ a die with a rotating dentate core and/or stator, so that a tubular net results. The tube or net may be slit to produce sheet. Processing is e.g. at 100-200°C.

In a yet further aspect the invention provides shaped articles made of the composition and compound structures comprising one or more such articles. Such shaped articles include substantially rigid articles such as vessels, pipes and pipe fittings, and flexible articles such as films, sacks and tubes of thickness typically in the range 0.001 to 1mm especially up to 0.1mm; and also nets of thread thickness typically in the range 0.01 to 1mm. Compound structures include especially combinations of such film, sack or tube or net with wood pulp, cellulose fibres or fibres of microbiologically produced copolymer such as PHB, PHBV and, indeed, compositions according to the invention. Particular structures include disposable diapers, tampons and wound dressings.

The invention may be further illustrated by reference to the following examples, in which percentage compositions are by weight unless otherwise stated and MI denotes the melt index in g/10 min according to ASTMD 1238.

EXAMPLE 1

A series of compositions was prepared comprising at least two of the following ingredients:

PHBV of 15 or 22 mol % 3-hydroxyvalerate.

EVA of VA content 19% w/w, MI 23 g/10 min ("MICROTHENE" MV 769-00 ex Quantum Chemicals).

EVA wax AC400 ex Allied Chemicals (viscosity 610 cps at 140°C).

LDPE of MI 6g/10 min ("MICROTHENE" MN72200 ex Quantum Chemicals).

The compositions were injection moulded into ASTM D356 impact and D638 tensile bars. Physical properties were determined approximately 1.5 months after moulding.

Flexural Modulus (3 point bend) represents the average value obtained from 4 replicates as determined on an Instron 1122, using a span length of 67 mm, and a crosshead speed of $5mm.min^{-1}$.

Tensile properties represent the average obtained from 5 replicates as determined on an Instron 1122, using a gauge length of 10 mm and a crosshead speed of 10 $mm.min^{-1}$.

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Izod impact strength (1 mm notch radius) represents the average obtained from 5 replicates as determined using a Zwick pendulum impact testing machine.

Weight loss is as determined on samples left in contact with microbiologically active soil for 79 days; under such conditions a sample of homopolymer of 3-hydroxybutyrate showed an 18% loss in weight.

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TABLE 1

LDPE %	EVA % (1)	PHBV %	HV (2)	FLEX. MOD.	YOUNGS MOD. (MPa)	STRESS @ BREAK (MPa)	EXTEN. @ BREAK %	IMPACT ($J.m^{-1}$)	WEIGHT LOSS %	Ó % (3)
100	0	0	0	265	84	19.5	133	*	-	0
100	0	0	0	245	82	21.8	164	*	-	-
100	0	0	0	285	77	18.2	148	*	-	-
60		40	22	588	203	15.4	22	43	-	-
50		50	22	668	230	15.9	19	38	-	-
60		40	15	499	195	9.4	25	63	10.9	5
70		30	15	441	169	9.4	35	78	9.13	-2
80		20	15	420	152	10.1	51	105	5.68	3.5
	70	30	20	213	58	2.4	564	208	-	-
40	30	30	20	377	121	3.5	125	141	-	-
40	30	30	20	330	102	0.3	283	188	-	-
50	20	30	20	363	119	7.4	61	94	-	-
20	40	40	22	403	122	1.9	97	85	-	-
30	30	40	22	306	121	6.6	61	64	-	-
50	20	30	15	333	121	7.4	70	112	-	-
60	10	30	15	395	149	10.1	60	86	-	-
30	40	30	15	233	62	5.3	91	240	1.7	0
30	30	40	15	278	93	5.0	72	60	5.2	0
30	30	40	15	263	93	5.9	49	64	-	-
40	20	40	15	347	128	7.1	56	55	5.96	5
40	20	40	15	326	122	8.0	49	51	-	-
50	10	40	15	421	159	9.6	39	47	8.46	4.5
	80	20	22	162	47	9.8	--	174	-	-
	40/20	40	22	403	122	1.9	97	85	-	-
60	10/10	20	15	337	104	9.1	130	266	-	-
60	10/10	20	22	304	102	10.2	85	245	-	-
20	20/20	40	22	258	91	7.2	36	38	-	-
20	30/10	40	22	348	112	7.3	38	50	-	-

(1) double entry indicates the percentages of EVA and EVA wax.

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(2) HV indicates the mol % of 3-hydroxyvalerate in the PHBV copolymer.

* - Sample failed to break.

(3) Indicates change in force at break.

It is evident that significant biodegeneration takes place, despite the presence of LDPE and/or EVA. However, it was observed also that exposure to soil caused only slight loss in mechanical strength.

EXAMPLE 2

The following polymer ingredients were used:

PHBV containing 14 mol % of HV units, with 10 phr of triacetin plasticiser;

EVA of MI 3.0 ml/10 min, 18% w/w VA. (CIL 1820)

LDPE CIL type 209

Various compositions % w/w were blown into films,by the conventional bubble method. Mechanical properties were measured and the biodegenerability of the films was assessed after 56 days' contact with a microbiologically active soil.

TABLE 2

LDPE %	EVA %	PHBV %	YIELD STGTH MPa	BREAK STGTH MPa	ELONG ACTION %	DENSITY KG M^{-3}	WEIGHT LOSS %	STGTH LOSS %
-	50	50	MD 9.5	15.4	479			
			TD 10.9	19.8	487	Over 1.0	41.7	23.1
-	75	25	MD 7.8	22.1	560			
			TD 6.4	14.2	476	995-1000	5.1	- 7.8
50	-	50	-	-	-	-	41.1	40.5
-	20	80	-	-	-	-	81.4+	100
100	-	-	9.0	14.0	650	919	0.0	-26
-	-	100	-	-	-	-	100.0	100

* MD = machine direction; TD - transverse direction

+ correction of copying error in priority application

It is evident that significant biodegradation, lending to loss in weight and strength, can be obtained if the content of PHBV is high enough. Thus material at under 50% PHBV could be used in making diapers, tampons and dressings to be disposed of as landfill.

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EXAMPLE 3

Using a PHBV sample containing 15.6 mol % HV and 1 phr boron nitride as nucleating agent, further compositions were formed into films and the biodegenerability assessed in a similar manner to Example 2, except that the time period was 79 days.

TABLE 3

EVA %	VA IN EVA MOL%	EVA MI	PHBV %	YIELD STGTH MPa	BREAK STGTH MPa	YOUNG'S MODULUS MPa	WEIGHT LOSS %
70	20	1.8	30	MD 7.4	7.9	290	15.5
				TD 4.3	5.2	140	
80	20	1.8	20	MD 5.8	6.5	201	2.67
				TD 4.5	4.9	79	
70	12	2.5	30	MD 9.55	11.7	327	11.6
				TD 5.8	6.0	177	
80	12	2.5	20	MD 7.5	10.6	222	2.22
				TD 4.4	5.5	112	

It is evident that significant weight loss occurs at the higher PHBV and EVA contents.

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EXAMPLE 4

Further compositions were injection moulded into impact and tensile test bars. The following physical properties were determined.

TABLE 4

EVA %	VA IN EVA	PHBV %	HV IN PHBV	FLEX. MOD. (MPa)	YOUNGS MOD. (MPa)	IMPACT ($J.m^{-1}$)
	(4)	100	4	2375	1170	92
10	8.5	90	4		1090	39
10	12.0	90	4		1050	36
10	20.0	90	4		1120	48
10	28.0	90	4		1080	57
30	8.5	70	4		778	41
30	12.0	70	4		734	44
30	20.0	70	4		711	52
30	28.0	70	4		642	67
50	8.5	50	4		524	58
50	12.0	50	4		487	60
50	20.0	50	4		480	89
50	28.0	50	4	619	396	193
		100	15.6	1207	642	197
10	8.5	90	15.6		533	65
10	12.0	90	15.6		558	69
10	20.0	90	15.6		554	84
10	28.0	90	15.6		509	132
30	8.5	70	15.6		404	52
30	12.0	70	15.6		414	56
30	20.0	70	15.6		397	76
30	28.0	70	15.6		362	107
50	8.5	50	15.6		300	77
50	12.0	50	15.6		271	93
50	20.0	50	15.6		247	139
50	28.0	50	15.6	302	202	292

(4) VA % w/w	8.5	12.0	20.0	28.0
MI g/10 min	2.0	2.5	1.8	3.0

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EXAMPLE 5

Further compositions were formed into impact and tensile test bars and the physical properties determined as in Example 4.

TABLE 5

EVA (5) %	PHBV %	HV IN PHBV	FLEX. MOD. (MPa)	YOUNGS MOD. (MPa)	NOTCHED IMPACT ($J.m^{-1}$)	WEIGHT LOSS %
	100	15.6	1082	642	250	24.01
10	90	15.6	867	465	151	-
20	80	15.6	619	374	101	-
30	70	15.6	462		117	23.6
40	60	15.6	457	273	304	22.35
50	50	15.6	323	51	461	17.75
60	40	15.6	232	132	367	10.36
70	30	15.6	156	80	287	4.38
80	20	15.6	97	50	210	1.5
100	0	-	47	17	-	-

(5) EVA containing 28% w/w of VA and having NI 3.0 g/10 min was used in all compositions in this example.

EXAMPLE 6

Further compositions, using PHBV comprising 15 mol % of HV and including some which contained other biodegradable materials apart from PHBV, were formed into tensile test bars and the physical properties determined as in Examples 4 and 5. Some of these compositions, as 0.5 mm compression moulded film, were tested for biodegradation for 30 days at 25°C in microbially active soil.

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Results were as follows.

TABLE 6

LDPE %	EVA (6) %	PHBV %	ST. (1) %	TRI. (2) %	Ca (3) %	BIO (4) %	POLY (5) %	FLEX. MOD. (MPa)	YOUNGS MOD. (MPa)	STRS @ MPa	EXTN BK %	WGT LOSS %
-	10a	50	-	20	20	90	56	344	204	7	27	45.1
-	10	40	-	25	25	90	44	379	193	-	-	-
-	10a	50	10	15	15	90	56	609	248	5	11	78.9
-	20a	40	10	15	15	80	50	345	134	-	-	-
-	40	30	-	15	15	60	50	291	132	-	-	-
-	40a	30	30	-	-	60	50	580	352	-	-	-
-	40	30	10	-	20	60	50	904	527	-	-	-
-	40a	30	20	5	5	60	50	458	211	-	-	-
-	50a	25	10	7.5	7.5	50	50	331	137	-	-	-
-	50	20	-	15	15	50	40	216	86	-	-	-
-	50	30	10	-	10	50	60	645	332	6	20	31.2
20	30	30	10	5	5	50	60	375	219	-	-	-
30	30	30	-	5	5	40	75	329	191	3	28	10.6
-	60	30	10	-	-	40	75	486	274	-	-	-
-	-	50	-	10	10x	60	83.3	605	345	14	16	43.3
-	-	56	-	10	10y	66	85	461	355	13	50	63.8

(1) ST. = starch

(2) TRI. = triacetin

(3) Ca. = calcium carbonate

(4) BIO = Biodegradable content of composition

(5) POLY = Percentage of biodegradable content which is PHBV.

(6) VA = 12% w/w in samples marked a; 8% in others. MI 2g/10mins for all EVAs.

(x) Contained also 30%) polycaprolactone "TONE" 700

(y) Contained also 24%) ex Union Carbide

All these samples were denser than fresh water.

The samples for which weight loss data are quoted had the following properties after the exposure to soil.

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TABLE 7

% WGT LOSS	COMMENTS
78.9	Broke into small pieces. Very fragile
45.1	Large amount of degradation - some fragmentation
31.2	Large amount of degradation - but still complete
10.6	Some degradation, but still complete
43.3	Degradation occurred at one end of strip
63.8	Significant degradation, broke into several pieces

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CLAIMS

1 A biodegenerable plastics composition comprising at least one synthetic polymer selected from the class consisting of polyethylene, polypropylene, polystyrene and an ethylene copolymer, blended with 10 to 90% w/w on said composition of a biodegradable material at least 20% w/w of which is microbiologically produced copolymer containing 3-hydroxybutyrate and 3-hydroxyvalerate.

2 A composition according to Claim 1 in which the ethylene the copolymer contains 5 to 30 w/w % of vinyl acetate on the total of ethylene and vinyl acetate.

3 A composition according to Claim 1 or Claim 2 containing also one or more biodegradable or environmentally acceptable improvers.

4 A composition according to any one of Claims 1 to 3 which comprises a ternary blend of

(1) said biodegradable material;

(2) at least one synthetic polymer selected from polyethylene, polypropylene and polystyrene; and

(3) said ethylene copolymer.

5 A composition according to any one of the preceding Claims in which the microbiologically produced copolymer contains between 3 and 20, especially between 5 and 15, mol % of 3-hydroxyvalerate.

6 A composition according to any one of the preceding claims having a density greater than that of fresh water.

7 A method of making a composition according to any one of the preceding claims by mixing the constituents in conditions in which at least one of its polymeric constituents is in liquid form.

8 A process of making a shaped article by confining to shaping surfaces a composition according to any one of Claims 1 to 6 or as made by a method according to Claim 7.

9 A shaped article made of a composition according to any one of Claims 1 to 6 or as made by a method according to Claim 7.

10 A compound structure comprising a shaped article according to Claim 9.

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INTERNATIONAL SEARCH REPORT

International Application No. PCT/US91/05245

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) [6]

According to International Patent Classification (IPC) or to both National Classification and IPC

IPC (5): C08J 3/02; C08K 5/09; C08L 23/00

U.S. CL. 523/124; 525/190

II. FIELDS SEARCHED

Minimum Documentation Searched [7]

Classification System	Classification Symbols
U.S.	523/124; 525/190

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched [8]

III. DOCUMENTS CONSIDERED TO BE RELEVANT [9]

Category *	Citation of Document, [11] with indication, where appropriate, of the relevant passages [12]	Relevant to Claim No. [13]
X	US, A, 3,929,937 (CLENDINNING ET AL.) 30 DECEMBER 1975; (see column 2, lines 40-61, column 6, lines 20-21, column 7, lines 36-38, and column 9, lines 23-31).	1-10
A	US, A, 4,900,299 (WEBB) 13 FEBRUARY 1990.	
A	US, A, 4,393,167 (HOLMES ET AL.) 12 JULY 1983.	
A	US, A, 4,620,999 (HOLMES) 04 NOVEMBER 1986.	

* Special categories of cited documents: [10]

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IV. CERTIFICATION

Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report
23 SEPTEMBER 1991	08 NOV 1991
International Searching Authority	**Signature of Authorized Officer**
ISA/US	Patrick Niland

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(54) Title: CHEMICALLY DEGRADABLE POLYOLEFIN FILMS

(57) Abstract

The present invention is directed to melt blown polyolefin film which is produced cheaply by melt blowing polyolefin resins which contain 25 to 60 wt% fillers selected from the fillers including an inorganic carbonate, synthetic carbonates, nepheline syenite, talc, magnesium hydroxide, aluminum trihydrate, diatomaceous earth, mica, natural or synthetic silicas and calcined clays or mixtures thereof, having a particle size less than 150 mesh. A further aspect of the present invention is directed to polyolefin resins, especially polyethylene, which are chemically degradable by the incorporation of the combination of the above group of fillers and a metal carboxylate. The metal carboxylates of the present invention are inclusive of a large number of metals, such as cerium, cobalt, iron, and magnesium.

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CHEMICALLY DEGRADABLE POLYOLEFIN FILMS

Field of the Invention

The present invention is directed to polyolefin resins, especially polyethylene resins, which are loaded at high levels with certain fillers to produce cheap polyolefin melt blown film products. More specifically, film made from polyolefins having both a filler such as an inorganic carbonate and a metal carboxylate are rendered chemically and thermally degradable. In addition, the present invention is directed to chemically and thermally degradable cast film polyolefin products which contain these certain fillers and a metal carboxylate.

The fillers of the present invention are preferably calcium carbonates, magnesium carbonate or other inorganic carbonates including the synthetic carbonates but include materials such as nepheline syenite, talc, magnesium hydroxide, aluminum trihydrate, diatomaceous earth, mica, natural or synthetic silica and calcined clays.

Background of the Invention

U.S. Patent 4,156,666 discloses degradable polyolefin resin compositions comprising the polyolefin resin, about 0.2 to 10% by weight of a fatty acid or esters of fatty acids with monohydric aliphatic alcohols and optionally, about 10 to 60% by weight of an inorganic filler such as calcium carbonate. These resins are clearly characterized as molding resins and the acid or esters are promoters for photodegradability. The degradation of the molding resins disclosed requires exposure to sunlight for degradation to occur.

It is well documented that the first issue regarding degradable plastics is the definition of "degradable". The classification of environmentally degradable materials was made by R. Narayan of Michigan Biotechnology Institute in "Degradable Materials: Perspectives, Issues and

Opportunities", pg.1, CRC Press, 1990, as "biodegradable, photodegradable and chemically degradable." The question was posed: "Are they (degradable plastics) a delusion, a solution or a downright hoax?" in an article by John Donnelly, "Degradable Plastics", Garbage, May/June 1990. As described in the article, the first attempts at producing a degradable polyolefin film, trash bags for example, was to add materials such as starch or sugar which made the plastic more biodegradable or to add materials such as metals which made the plastic more photodegradable. However, each of these materials were dependant on the environment of the plastic; for example the presence of bacteria for the biodegradable reaction to occur or sunlight (UV) for the photodegradable reaction to occur.

The present invention is directed to a combination of additives which produce a chemical degradation of the plastic over a period of time. The compounding of the additives to the plastic resins permits the desired use, trash bag or agricultural film, but will by chemical reaction cause the plastic product to be environmentally degradable. This chemical reaction is accelerated by heat and/or UV light but will degrade without sunlight when heated to temperatures such as found when the plastic is buried in landfills.

Summary of the Invention

The present invention is directed to melt blown polyolefin film which is produced cheaply by melt blowing polyolefin resins which contain 25 to 60 wt% fillers selected from the fillers including an inorganic carbonate, synthetic carbonates, nepheline syenite, talc, magnesium hydroxide, aluminum trihydrate, diatomaceous earth, mica, natural or synthetic silicas and calcined clays or mixtures thereof, having a particle size less than 150 mesh.

A further aspect of the present invention is directed to polyolefin resins, especially resins containing polyethylene, which are chemically degradable by the incorporation of the

combination of the above group of fillers and a metal carboxylate. The metal carboxylates of the present invention are inclusive of a large number of metals, such as cerium, cobalt, iron, and magnesium.

Brief Description of the Preferred Embodiments

The present invention is directed to melt blowing polyolefin resins which contain 25 to 60 wt% of certain fillers. Heretofore, polyolefin resins used in melt blowing did not contain over 10 wt% of any filler since the fillers in most film grade resins would cause tearing or holes in the film. According to the present invention it was found that certain polyolefin resins can be used to produce melt blown film containing certain fillers at levels heretofore not thought possible. The fillers which can be used to fill a polyolefin resin are selected from the inorganic carbonate, synthetic carbonates, nepheline syenite, talc, magnesium hydroxide, aluminum trihydrate, diatomaceous earth, mica, natural or synthetic silicas and calcined clays or mixtures thereof, having a particle size less than 150 mesh. It was further found that these fillers needed to be free of water. The surface of these fillers are treated so as to not adsorb water which will produce steam and holes when melt blown into film. Accordingly, these fillers, such as calcium carbonate, are treated with organic acids to assist the processability of the carbonate and produce a more hydrophobic filler product. Acids such as stearic or oleic acid are conventional acids for surface treating the carbonates or other fillers. The surface treatment is usually done by the carbonate suppliers and a surface treated filler is part of the present invention.

The inorganic carbonates such as calcium carbonate or magnesium carbonate are preferred as fillers; however, lithium carbonate, sodium carbonate or sodium bicarbonate may also be used. In addition, the synthetic carbonates such as the hydrotalcite-like compound or the dihydroxyaluminium

sodium carbonates may be used in the present invention. In addition to the inorganic or synthetic carbonates, a filler such as nepheline syenite, talc, magnesium hydroxide, aluminum trihydrate, diatomaceous earth, mica, natural or synthetic silicas including silicon dioxide and calcined clays or mixtures thereof, having a particle size less than 150 mesh may be used. All the fillers of the present invention have particle sizes less than 150 mesh but the smaller the particle size of the filler material, the more preferred it is as the filler material. The most preferred filler is calcium carbonate having a 1 to 10 micron particle size.

The polyolefins which are melt blown into film products according to the present invention include low density polyethylene (LDPE), linear low density polyethylene (LLDPE), polypropylene, polybutylene and copolymers of ethylene such as polyethylene-vinyl acetate (EVA), polyethylene-acrylic acid (EAA), polyethylene-methacrylic acid (EMA) or copolymers of ethylene or propylene with the lower olefins such as, butene-1, pentene-1, hexene or octene. The term "polyethylene resin" as used herein includes any resin where ethylene is predominant and is illustrated by the polyethylene resins in the foregoing list.

The polyolefin resins which are capable to be melt blown at filler levels of 15 to 65 wt%, preferably 20 to 55 wt%, have the characteristic that they are usually copolymers of ethylene or propylene. A common characteristic of the olefin resins used in the present invention are resins having high elongations (having an ultimate elongation {ASTM-D-638} of over 500%) and high melt strength which is usually associated with the property of high elongation. The copolymer resins usually have a significant amount (over 2 wt%, such as 2-25 wt%) of co-monomer and include the block copolymer resins. One group of polyolefin resins are those copolymers of ethylene which include acrylic acid, methacrylic acid or vinyl acetate or monomers which have the characteristic of

highly flexible thermoplastics. Another group of polyolefin resins are those which include ethylene or propylene and other olefins such as butene, hexene or octene as co-monomers to produce the copolymers. The term "polyolefin copolymer resin" as used herein includes those polyethylene or polypropylene resins which have at least one other co-monomer. The density of the polyethylene resins are between 0.70 and 0.97 and the molecular weights may range between 100,000 and 250,000. The melt index of these polyethylene resins may range from 20 to about 1 (gram/minute ASTM D 1238). The range of melt index is broad for applicable resins but it is not intended to infer that this broad range is applicable to each specific resin, but only that resins from that range of melt index when filled with the fillers of the present invention can be melt blown into film. These polyolefin resins used in the present invention are not resins which are necessarily produced and sold as film grade resins. However, when the high loading of the fillers of the present invention are incorporated into these polyolefin resins, the compounded resins are capable of being melt blown as a film.

Polyolefin resins that are injection molding resins or are unsuitable for the present invention are characterized as having small ultimate elongations, usually 100 to 300%, with no melt strength.

Melt blowing of polyolefins is a well known and established process for making polyolefin film; however, melt blowing films which contain 10 wt%, or more specifically greater than 20 wt% of a filler has not been done heretofore. Surprisingly, these high loadings of fillers is without detriment to the film blowing capabilities of the polyolefin resins of the present invention. In the melt blowing process, the polyolefin is extruded through a circular die having a gas, usually air, blown into the circle of resin to form a bubble which is pulled upward until the resin cools and the resin film is then run through rollers which collapse

the bubble and forms a continuous cylinder of film. The apparatus for melt blowing film consists of an extruder, a circular die, an air ring for blowing air into the die, a collapsible frame for collapsing the bubble formed, nip rolls and a winder for the collapsed film. The film is then slit to form a sheet which usually is twice the size of the collapsed film (a cylinder of film) or in other words is about the size of the circumference of the bubble formed.

The present invention is further directed to chemically degradable polyolefin melt blown film. According to the present invention, melt blown polyolefin films are chemically degradable when a certain chemical prodegradant, specifically a metal carboxylate, is combined with the foregoing certain fillers. The degradable resins of the present invention are produced by incorporating the combination of certain fillers, which may be mixtures of fillers, and the chemical prodegradant, a metal carboxylate, to resins which are able to be melt blown into film and mixing the filler and prodegradant in an extruder and then forming the pellets which are used in the conventional melt blown film equipment. The term "degradable" as used herein means that the polyolefin exhibits more than loss of physical properties such as brittleness or lost of tensile strength but loss of molecular weight. The degradation of the polyolefin resins is clearly characterized as chemical since the degradation does not require UV light (photodegradable) or a bio initiator (biodegradable) for the degradation to be initiated and to take place. On the other hand, UV light (sunlight) will accelerate chemical degradation. Unique to the present invention is that the degradation additives, the combination of the fillers and the prodegradant, namely, the carbonate and metal carboxylate, will exceed 10 wt%, and can be greater than 20 wt%, and can be as high as 45 wt% or higher and formed into films of less than 1 to up to 25 mil or greater.

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The prodegradant additives of the present invention which are combined with the fillers to produce the degradable products of the present invention are the metal carboxylates. The preferred metal carboxylates are cobalt, cerium and iron stearate. Other suitable metal carboxylates are carboxylates containing aluminum, antimony, barium, bismuth, cadmium, chromium, copper, gallium, lanthanum, lead, lithium, magnesium, mercury, molybdenum, nickel, potassium, rare earths, silver, sodium, strontium, tin, tungsten, vanadium, yttrium, zinc or zirconium.

A chemically degradable polyolefin film resin may be produced with as little as one weight percent (1 wt%) of each additive, 1 wt% of the selected carbonate fillers and 1 wt% of a metal carboxylate. However, it has been found that it is most desirable that the fillers of the present invention are added at high levels (10 wt% to 45 wt%) to the film producible polyolefin resins to produce a chemically degradable polyolefin film. The range of filler may be between 1 and 45 wt% and preferably between 5 and 25 wt% depending on the use of the film. For example, an agricultural film that requires the film integrity and the physical characteristics of the film for a short period of time will have a higher loading or greater weight percentage of filler than a film used in bags where the strength of the film may be required for a longer period of time.

The amount of metal carboxylate incorporated into the degradable film grade resins of the present invention is between 0.1 and 15 wt%, preferably between 0.5 and 3 wt%. Only a small amount of metal carboxylate may be required and amounts less than 1 wt% may be suitable, especially at very high loadings of fillers. The amount of metal carboxylate has a much greater effect on the rate of degradability than the amount of filler; however, one effect that the high loading of filler has is to reduce the total wt% of plastic present. Thus, the rate of degradability of the olefin plastic may be increased by increasing the amount of metal

carboxylate or substantially increasing the amount of fillers. Another factor, however, is the environment of polyolefin film product which will accelerate the degradation process; hence, if the product is used outdoors and subjected to sunlight (UV), for example, the total degradation of the film product is accelerated. However, unlike compositions of the past, the prodegradant does not require UV light to initiate the degradation. Heat alone initiates the chemical degradation reaction, temperatures in excess of 100 °F and especially in excess of 120 °F will initiate the degradation especially in the presence of oxygen.

The degradable melt blown films of the present invention have uses in several different markets and depending on the market may contain additives which will have special application for that market. One market for use of the degradable films is as a landfill cover. Each night the open area or face, the area where trash has been dumped during operation, of the landfill requires a covering, which presently uses a six (6") inch covering of dirt, sand or other material. A significant advantage of the films of the present invention is that a reduction of 156 feet/year (6" x 6 days x 52 weeks) in the height of the landfill is possible. Since melt blown polyolefin films do not use a plasticizer or solvent, the film does not give off solvents or odors. Another advantage is that the film is chemically degradable under the conditions of compacting or multiple layers of trash placed in the landfill which raises the temperature to levels where the films of the present invention will degrade by a chemical/thermal reaction. The additives which are used in the landcover films are primarily color additives to provide a wide range of color choices. In addition, to overcome the usual bad odors of a landfill, an additive which provides a favorable smell can be added to the films of the present invention.

Another market for the degradable films of the present invention is agricultural films which conserves moisture and

prevents weeds from growing, taking the nutrients from the growing crop. These polyolefin film covers provide protection from pests, wind and moisture loss; and cut down or eliminate the need for chemical sprays. The additives added to an agricultural film are in addition to color, nutrients such as trace elements and fertilizers. When the film degrades, these trace elements and fertilizers will aid the growth of the crop. Similar to the large scale agricultural market is the home garden market where the films of the present invention will contain the trace elements and fertilizers.

Still a further market is the conventional film markets; however, what is added to the film is the low cost and degradability features of the present invention. Such products as films for packaging, wraps, containers and disposables, especially disposable diapers and bags. In these applications, additional additives are not required but FDA approved materials may be added to modify properties.

As noted hereinabove, it is common practice that the commercial calcium carbonates will be surface treated with 1 wt% of an organic acid such as stearic acid or oleic acid before adding with the fillers to aid the processability of the carbonate but because the amounts are at most about 1 wt%, they are not separately accounted for in the formulations hereinafter.

The following examples, all formulations or recipes given in weight percent (wt%), illustrate some resins, the fillers and metal carboxylates used in the present invention:

EXAMPLE 1

60 wt% of a polyethylene - 4% vinyl acetate (EVA) resin (PE 5272, a product of Chevron Chemical Company); and 40 wt% calcium carbonate ($CaCO_3$) are mixed and added to an extruder having a barrel temperature range of about 300-425 °F. and the strands pelletized to produce a film grade resin. This film grade resin is melt blown into

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a film of about 2 mil thickness and illustrates the melt blowing of a polyolefin resin with very high filling.

This example illustrates that very high loading of fillers in a polyolefin resin will produce a melt blown polyolefin film.

EXAMPLE 2

50 wt% polyethylene- vinyl acetate (EVA) copolymer (Elvax 260, a product of DuPont Chemical Company); and

50 wt% calcium carbonate are compounded, pelletized and blown into a 1 to 5 mil film.

The polyethylene (EVA) resin of this example has a density of 0.948 (ASTM D 1505), a melt index of 6 MI (ASTM 1238, modified). Even at the high loading level of 50 wt% of filler, a film is melt blown and surprisingly the line speeds can be increased since the film is easily cooled in that the fillers act as a heat sink. The addition of a filler which costs under $0.10/lb when added to resins which at their cheapest are about $0.30/lb and may be as much as $0.75/lb, illustrates the advantage of the present invention.

EXAMPLE 3

65 wt% polypropylene (HGZ 350, a product of Phillips Chemical company)

35 wt% calcium carbonate (Kodomite, a product of Thompson Weiman) are compounded, pelletized and cast into a 1 to 5 mil film.

It is clear that according to the present invention, film is produced from polyolefin resins having incorporated therein certain fillers, primarily polyethylene resins, to produce low cost film, especially suitable for disposable film products. Heretofore, the addition of additives to the melt blown polyolefins was solely the usual stabilizers, antiblock or carbon black or other coloring additives and were at low levels, usually less than 10 wt% and more normally less than 5 wt%. The levels of fillers selected from calcium carbonates, magnesium carbonate or other

inorganic carbonates but which may include materials such as nepheline syenite, talc, magnesium hydroxide, mica, natural or synthetic silicas and calcined clays are between 10 to 60 wt% of the fillers and preferable between 15 to 50 wt%, with levels between 35 and 45 wt% of calcium carbonate easily producing a melt blown film. The fillers all have particle sizes less than 150 mesh and preferably sizes between 5 and 30 microns. The high loaded resins of the present invention may be melt blown into a 1 to 10 mil film.

The following Examples illustrate degradable films by the simple addition of a metal carboxylate. Other selected additives may be included in the specific films.

EXAMPLE 4

60 wt% of a polyethylene - 4% vinyl acetate (EVA) resin (PE 5272, a product of Chevron Chemical Company);

39 wt% calcium carbonate ($CaCO_3$), acid coated; and

1 wt% cobalt stearate are mixed and added to an extruder having a barrel temperature range of about 300-425 °F. and the strands pelletized to produce a film grade resin. This example illustrates the combination of filler and a metal carboxylate. The compounded resin is melt blown into a degradable film.

This example illustrates a degradable resin for melt blowing agricultural film which after a short period of time under high temperature conditions will degrade.

EXAMPLE 5

55 wt% of a ethylene - octene copolymer (2045, a product of Dow Chemical Company);

40 wt% calcium carbonate ($CaCO_3$);

3 wt% cerium stearate; and

2 wt% carbon black is mixed and added to an extruder having a barrel temperature range of about 300-425 °F. and the strands pelletized to produce a film grade

degradable resin. The compounded resin is melt blown into a degradable film.

The foregoing example illustrates the combination of the prodegradant, cerium stearate, a filler, a carbonate and other fillers such as carbon black which will color the film.

The following example illustrates a formulation with a combination of fillers.

EXAMPLE 6

50 wt% of a ethylene-vinyl acetate (EVA) copolymer (ELVAX 350, a product of DuPont Chemical Company, 25 % VA and 19 MI);

33 wt% calcium carbonate ($CaCO_3$);

10 wt% talc;

5 wt% nepheline syenite (Minex 7, a product of Unimin Canada Limited)

2 wt% cobalt stearate is mixed and added to an extruder having a barrel temperature range of about 300-425 °F. and the strands pelletized to produce a film grade degradable resin. The compounded resin is melt blown into a degradable film.

The following examples include more than one metal carboxylate combined with the fillers.

EXAMPLE 7

60 wt% of a polyethylene - 4% vinyl acetate (EVA) resin (PE 5272, a product of Chevron Chemical Company);

38 wt% calcium carbonate ($CaCO_3$);

1 wt% cerium stearate; and

1 wt% cobalt stearate is mixed and added to an extruder having a barrel temperature range of about 300-425 °F. and the strands pelletized to produce a film grade degradable resin. The compounded resin is melt blown into a degradable film.

EXAMPLE 8

50 wt% of a ethylene-hexene copolymer (VLLDPE 1539, a product of Union Carbide Company);

48 wt% calcium carbonate ($CaCO_3$);

1 wt% iron stearate; and

1 wt% cerium stearate is mixed and added to an extruder having a barrel temperature range of about 300-425 °F. and the strands pelletized to produce a film grade degradable resin. The compounded resin is melt blown into a degradable film.

The following example illustrates that more than one resin can be used to produce the degradable polyolefin resin of the present invention.

EXAMPLE 9

30 wt% of a ethylene-hexene copolymer (VLLDPE 1539, a product of Union Carbide Company);

30 wt% of a polyethylene - 4% vinyl acetate (EVA) resin (PE 5272, a product of Chevron Chemical Company);

38 wt% calcium carbonate ($CaCO_3$);

1 wt% cobalt stearate; and

1 wt% iron stearate is mixed and added to an extruder having a barrel temperature range of about 300-425 °F. and the strands pelletized to produce a film grade degradable resin. The compounded resin is melt blown into a degradable film.

This example illustrates the flexibility obtained by using two different resins to obtained desired film characteristics and two different prodegradants which have differing degradation rates.

EXAMPLE 10

30 wt% of a propylene-ethylene copolymer (HIMONT KS051, a Catalloy-process resin product of Himont, Inc.);

30 wt% of a polyethylene - 4% vinyl acetate (EVA) resin (PE 5272, a product of Chevron Chemical Company);

37 wt% calcium carbonate ($CaCO_3$); and

3 wt% cobalt stearate is mixed and added to an extruder having a barrel temperature range of about 300-425 °F. and the strands pelletized to produce a film grade degradable resin. The compounded resin is melt blown into a 4 mil degradable film.

EXAMPLE 11

To the extruder of a melt blowing apparatus, the compounded resins of Example 10 is added together with equal amounts of the polyethylene - 4% vinyl acetate (EVA) resin (PE 5272, a product of Chevron Chemical Company). The mixed resins thus have a 18.5 wt% of calcium carbonate and 1.5 wt% of the cobalt stearate. The compounded mixed resin is melt blown into 1 mil, 5 mil and 8 mil film.

EXAMPLE 12

The control resin is a linear low density polyethylene (LLDPE 7510, a product of Union Carbide).

A first sample is prepared:

57.5 wt% linear low density polyethylene (7510, a product of Union Carbide);

40 wt% calcium carbonate; and

2.5 wt% cobalt stearate are compounded, pelletized and blown into a 1 to 5 mil film.

A second sample is prepared:

55 wt% of a mixture of linear low density polyethylene and polypropylene (60 wt% LLDPE and 40 wt% PP);

40 wt% calcium carbonate;

2.5 wt% carbon black; and

2.5 wt% cobalt stearate are compounded, pelletized and blown into a 1 to 5 mil film.

Each sample is exposed to a UV lamp for 30 days. The molecular weight of the samples is found (source: LARKS ENTERPRISES of Webster, MA) as shown in Table 1:

Table 1

SAMPLE	MOL. WT. NO. AVE.	MOL. WT. WT. AVE.	Dispersity
Control	49490	242420	4.90
	49502	242492	4.90
	50293	243080	4.83
Control ave	49762	242664	4.88

SAMPLE	NO. AVE.	WT. AVE.	Dispersity
1st	2789	7920	2.84
	2688	7741	2.88
	2743	7859	2.87
1st ave	2740	7840	2.86
2nd	2192	4240	1.93
	2121	4089	1.93
	2067	4120	1.99
2nd ave	2127	4150	1.95

From the foregoing data it is shown that the degradation of the polyolefins containing the combination of both a filler, calcium carbonate, and a metal carboxylate, cobalt stearate, degrade in the accelerated test to a waxy material. Thus, there is more than a reduction of molecular weight but a chemical degradation breaking the polymer into small molecules.

The same degradation is expected in the formulations in Examples 4-11, i.e., the samples will result in waxy material having melt indexes that are so high that they are difficult to measure.

Samples of films illustrated in Example 4-12 degrade when placed outside in about 1 to 2 weeks or 1 to 6 months depending on the specific levels of fillers or metal carboxylates and/or type of resin used. The samples also show degradation, loss of molecular weight and much greater melt indexes, by heating samples at temperatures of about 130 °F. Clearly the degradation of the formulations of the present invention is chemical in nature and is initiated by either UV light or heat in the absence of sun light, typical of landfill conditions.

A specific application for the degradable resins of the present invention is agricultural film. Such film is used in agriculture as a cover to prevent weeds and other growth between the desired plants, such as onions, corn or tomatoes.

EXAMPLE 13

A formulation of Example 4 is melt blown as a 3-4 mil sheet and when placed outdoors degrades in about four to six weeks.

EXAMPLE 14

An agricultural film formulation similar to Example 4 is changed by adding small amounts, less than 1 wt% of trace metals such as manganese, copper, zinc, iron or nitrogen containing materials such as ammonium polyphosphate which when the film degrades enhances the soil or growing conditions of the desired plants.

EXAMPLE 15

Another agricultural mulch film formulation:

50 wt% polyethylene- vinyl acetate (EVA) copolymer

(Elvax 470, a product of DuPont Chemical Company);
35 wt% calcium carbonate;
1.5 wt% cobalt stearate;
8.5 wt% ammonium polyphosphate; and
5 wt% trace elements (Mg, Cu, Zn, Fe).

EXAMPLE 16

The addition of polypropylene resin to the formulation with polyethylene enhances the degradation. A typical formulation containing polypropylene is:

53 wt% polyethylene - 4% vinyl acetate (EVA) resin (PE 5272, a product of Chevron Chemical Company)

15 wt% polypropylene (HGZ 350, a product of Phillips Chemical company)

31.5 wt% calcium carbonate (Kodomite, a product of Thomson Weiman); and

0.5 wt% cobalt stearate.

The compounded resin is melt blown into a degradable film. The 1 to 5 mil film from this formulation degrades outside in about 1 to 6 weeks.

EXAMPLE 17

Formulations are prepared with other polyolefins as follows:

69 wt% polyolefin resin (the numbers in parenthesis { } are the ultimate elongations of the resin);
20 wt% calcium carbonate;
9 wt% nepheline syenite;
0.5 wt% cerium stearate;
0.5 wt% cobalt stearate;
1 wt% carbon black

using the following polyolefins:

polyethylene-acrylic acid (EAA resin 435 {650%}; 452 {570%}; 455 {650%}; or 459 {650%}, products of Dow Chemical Company);

polyethylene-octene copolymers (ATTANE 4001 {1000%}; ATTANE 4002 {1000%}; or ATTANE 4003 {760%}, trademarks and products of Dow Chemical Company);

linear low density polyethylene (DOWLEX 2045 {1000%}, a product of Dow Chemical Company);

polyethylene-vinyl acetate (ELVAX 470 {750%}, trademark and products of DuPont Chemical Company)

polyethylene-methylacrylic acid (NUCREL 925 {520%}, trademark and products of DuPont Chemical Company)

polyethylene- 20% methyl acrylate (PE 2260 {800%}, a product of Chevron Chemical Company)

polyethylene-butene copolymer (LLDPE-7510 NATURAL {800%}, a product of Union Carbide).

polyethylene-hexene copolymer (VLLDPE-1539 {900%}, a product of Union Carbide).

This list of resins includes acidic resins such as the EAA resins, the ELVAX resins, the NUCREL resins and the methyl acrylate resins which when combined with the carbonates become degradable due to the reaction of the acid and the base to form carbon dioxide and water.

The invention is further illustrated by blown films of more conventional polyethylene film grade resins which are made degradable by the inclusion of the metal carboxylate.

EXAMPLE 18

91.75 wt% of a polyethylene resin (PE 5272, a product of Chevron Chemical Company);

7.5 wt% calcium carbonate ($CaCO_3$), acid coated; and

0.75 wt% cobalt stearate are mixed and added to an extruder having a barrel temperature range of about 300-425 °F. and the strands pelletized to produce a film grade resin. The compounded resin is melt blown into a degradable film. This example illustrates the combination of filler and a metal carboxylate.

EXAMPLE 19

91 wt% of a polyethylene resin (PE 5272, a product of Chevron Chemical Company);

7.5 wt% calcium carbonate ($CaCO_3$), acid coated; and

1.5 wt% cobalt stearate are mixed and added to an extruder having a barrel temperature range of about 300-425 °F. and the strands pelletized to produce a film grade resin. The compounded resin is melt blown into a degradable film. This example illustrates the combination of filler and a metal carboxylate.

All of the foregoing examples included polyolefins as part of the formulation which are melt blown into degradable films.

The present invention also includes a cast polyolefin film which is degradable. Heretofore, high loading of cast film resins have not been considered. However, it has been found that a cast film which is degradable by loading with the fillers and metal carboxylates of the present invention.

When the polyolefin resins or compounds are designed for cast film applications, the molecular weight distribution is critical. In order to extrude a smooth surface at high extrusion rates the molecular weight distribution must be narrow. Molecular weight distribution can be measured as a ratio of HLMI (ASTM 1238) to regular MI. The ratio of HLMI to MI when less than 6 indicates a narrow molecular weight distribution and such resins are used to produce the cast polyolefin films of the present invention.

The present invention of a degradable polyolefin film is illustrated by a formulation for a cast film.

EXAMPLE 20

37 wt% of a ethylene-hexene copolymer (VLLDPE 1539, a product of Union Carbide Company);

30 wt% polypropylene (HGZ 350, a product of Phillips Chemical Company);

30 wt% calcium carbonate;

2 wt% cerium stearate;

1 wt% titanium dioxide

This formulation is cast as a film.

The foregoing examples are illustrative of the present invention and the ranges and specific additives may be changed. The addition of additives in polyethylene resins may be as great as 60 wt% ; whereas, with polypropylene resins the loading may be up to 50 wt%.

Some of the fillers, especially the synthetic silicas, can be tailored to have a refractive index which is essentially the same as the polyolefin resin or can be modified by the size of the particle to make the refractive index the same as the resin. When the refractive index of the filler and the resin is the same a clear film is produced. Some of the prodegradants such as cerium stearate are also clear which permits the first clear degradable film to be produced.

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Claims:

1. A process for melt blowing a polyethylene resin comprising: extruding a composition comprising 40 to 75 wt% polyethylene resin and 60 to 25 wt% calcium carbonate having a particle size between 0.5 and 10 microns through a melt blowing die; and blowing a bubble of said composition which is collapsed into a film.

2. A process for melt blowing a polypropylene resin comprising: extruding a composition comprising 50 to 65 wt% polypropylene resin and 50 to 35 wt% calcium carbonate having a particle size between 0.5 and 10 microns through a melt blowing die; and blowing a bubble of said composition which is collapsed into a film.

3. A process for melt blowing a polyolefin resin comprising: extruding a polyolefin composition comprising 40 to 90 wt% of a polyolefin resin and 60 to 10 wt% of a filler selected from the group consisting of an inorganic carbonate, synthetic carbonates, talc, magnesium hydroxide, mica, natural or synthetic silicas and calcined clays, having a particle size less than 150 mesh through a melt blowing die; and blowing a bubble of said composition which is collapsed into a film.

4. A process according to claim 3 wherein the refractive index of said filler is essentially the same as that of said resin such that a clear film is produced.

5. A 1 - 10 mil film of polyolefin resin produced from the process of claim 3.

6. A process for melt blowing a degradable polyolefin resin comprising: extruding a polyolefin composition comprising 55 to 98 wt% polyolefin resin; 30 to 1 wt% a filler selected from the group consisting of an inorganic carbonate, synthetic carbonate, talc, magnesium hydroxide, mica, natural or synthetic silicas and calcined clays, having a particle size less than 150 mesh; and 15 to 1 wt% metal carboxylate through a melt blowing die; and blowing a bubble of said composition which is collapsed into a film.

7. A 1 - 10 mil film of polyolefin resin produced from the process of claim 6.

8. A process according to claim 6 wherein: said polyolefin is polyethylene resin; and contains 25 to 1 wt% filler; and 10 to 1 wt% metal carboxylate.

9. A process according to claim 6 wherein the components are: at least 55 wt % and up to 88 wt% polyolefin resin; 43 to 10 wt% filler; and 10 to 1 wt% metal carboxylate.

10. A degradable polyolefin composition consisting essentially of: 35 to 89 wt% polyolefin resin having an ultimate elongation in excess of 500%; 10 to 50 wt% a filler selected from the group consisting of an inorganic carbonate, talc, magnesium hydroxide, mica, natural or synthetic silicas and calcined clays, having a particle size less than 150 mesh; and 1 to 15 wt% metal carboxylate.

11. A degradable composition according to claim 10 wherein: said polyolefin is a polyethylene resin; and contains 15 to 25 wt% filler; and 1 to 10 wt% metal carboxylate.

12. A degradable composition according to claim 10 wherein the components are: at least 55 wt % and up to 89 wt% polyolefin resin; 10 to 45 wt% filler; and 1 to 10 wt% metal carboxylate.

13. A degradable composition according to claim 10 wherein the ratio of HLMI to MI of the polyolefin resin is less than 6.

14. A polyethylene melt blown film grade composition comprising: 40 to 75 wt% polyethylene resin having an ultimate elongation in excess of 500%; and 60 to 25 wt% calcium carbonate having a particle size between 0.5 and 10 microns.

15. A 1 - 10 mil film of polyethylene produced by melt blowing the composition of claim 14.

16. A polypropylene film grade composition comprising: 50 to 65 wt% polypropylene resin; and 50 to 35 wt% calcium carbonate having a particle size between 0.5 and 10 microns.

17. A polyolefin film grade composition comprising: 40 to 90 wt% of a polyolefin resin having an ultimate elongation in excess of 500%; and 60 to 10 wt% of a filler selected from the group consisting of an inorganic carbonate, synthetic carbonate, talc, magnesium hydroxide, mica, natural or synthetic silicas and calcined clays, having a particle size less than 150 mesh.

18. An agricultural polyolefin film grade composition according to claim 17 comprising: 40 to 89 wt% polyolefin resin; 50 to 10 wt% filler; and 10 to 1 wt% of a material selected from the group consisting of fertilizers and trace metals.

19. A composition according to claim 17 wherein the refractive index of said filler is essentially the same as that of said resin such that a clear film may be produced.

20. A degradable polyolefin composition comprising: 55 to 98 wt% a polyolefin resin having an ultimate elongation in excess of 500%; 30 to 1 wt% a filler selected from the group consisting of an inorganic carbonate, synthetic carbonate, talc, magnesium hydroxide, mica, natural or synthetic silicas and calcined clays, having a particle size less than 150 mesh; and 15 to 1 wt% metal carboxylate.

21. An agricultural degradable polyolefin composition according to claim 20 comprising: 55 to 97 wt% polyolefin resin; 20 to 1 wt% filler; 15 to 1 wt% metal carboxylate; and 10 to 1 wt% of a material selected from the group consisting of fertilizers and trace metals.

22. A degradable film grade composition according to claim 20 wherein: said polyolefin is polyethylene resin; and contains 25 to 1 wt% filler; and 10 to 1 wt% metal carboxylate.

23. A polyethylene melt blown film comprising: 40 to 75 wt% polyethylene resin having a density of 0.70 to 0.97

and a molecular weight of between 100,000 and 250,000; and 60 to 25 wt% calcium carbonate having a particle size between 0.5 and 10 microns.

24. A polypropylene melt blown film comprising: 50 to 65 wt% polypropylene resin; and 50 to 35 wt% calcium carbonate having a particle size between 0.5 and 10 microns.

25. A polyolefin melt blown film comprising: 40 to 90 wt% of a film grade polyolefin resin; and 60 to 10 wt% of a filler selected from the group consisting of an inorganic carbonate, talc, magnesium hydroxide, mica, natural or synthetic silicas and calcined clays, having a particle size less than 150 mesh.

26. A film according to claim 25 wherein the refractive index of said filler is essentially the same as that of said resin such that said film is clear.

27. Compounding a polyolefin composition for producing a film grade resin which comprises: mixing 40 to 90 wt% of a polyolefin resin having an ultimate elongation in excess of 500% with 60 to 10 wt% of a filler selected from the group consisting of an inorganic carbonate, synthetic carbonate, talc, magnesium hydroxide, mica, natural or synthetic silicas and calcined clays, having a particle size less than 150 mesh; extruding said mixture into strands; and pelletizing said strands to produce a film grade resin.

28. Compounding a polyolefin composition for producing a film grade resin which comprises: mixing 55 to 98 wt% a polyolefin resin having an ultimate elongation in excess of 500% with 30 to 1 wt% a filler selected from the group consisting of an inorganic carbonate, synthetic carbonate, talc, magnesium hydroxide, mica, natural or synthetic silicas and calcined clays, having a particle size less than 150 mesh and 15 to 1 wt% metal carboxylate; extruding said mixture into strands; and pelletizing said strands to produce a film grade resin.

29. A method for covering landfills which comprises: dumping trash into a landfill during a period of operation

creating an open face of trash; rolling a film of degradable polyolefin resin over the open face of the landfill; and placing additional trash over the polyolefin film during a further period of operation.

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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 5 : **C08K 3/00, C08L 23/02, C08K 5/09 E02D 31/00 // (C08K 3/00, 3:22, 3:26, 3:36) (C08L 23/02, 23:06, 23:12)**	**A3**	**(11) International Publication Number:** **WO 94/04606** **(43) International Publication Date:** 3 March 1994 (03.03.94)

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104,195	10 August 1993 (10.08.93)	US

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(74) Agent: CHRYSTAL, John, J.; Ladas & Parry, 224 S. Michigan Ave., Chicago, IL 60604 (US).

(81) Designated States: CA, JP, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

Published
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Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.

(88) Date of publication of the international search report:
09 June 1994 (09.06.94)

(54) Title: CHEMICALLY DEGRADABLE POLYOLEFIN FILMS

(57) Abstract

The present invention is directed to melt blown polyolefin film which is produced cheaply by melt blowing polyolefin resins which contain 25 to 60 wt% fillers selected from the fillers including an inorganic carbonate, synthetic carbonates, nepheline syenite, talc, magnesium hydroxide, aluminum trihydrate, diatomaceous earth, mica, natural or synthetic silicas and calcined clays or mixtures thereof, having a particle size less than 150 mesh. A further aspect of the present invention is directed to polyolefin resins, especially polyethylene, which are chemically degradable by the incorporation of the combination of the above group of fillers and a metal carboxylate. The metal carboxylates of the present invention are inclusive of a large number of metals, such as cerium, cobalt, iron, and magnesium.

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INTERNATIONAL SEARCH REPORT

Inter nal Application No
PCT/US 93/07922

A. CLASSIFICATION OF SUBJECT MATTER
IPC 5 C08K3/00 C08L23/02 C08K5/09 E02D31/00 //(C08K3/00, 3:22,3:26,3:36),(C08L23/02,23:06,23:12)

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 5 C08K

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 practical, 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	DATABASE WPI Week 7809, Derwent Publications Ltd., London, GB; AN 75-44981W & JP,A,49 104 956 (UBE INDUSTRIES) 4 October 1974 see abstract ---	1,3,23
X	US,A,5 064 893 (M.F. HOENIGMANN) 12 November 1991 see column 1, line 13 - line 16 see column 1, line 49 - line 53 see claims 1-8; examples; table 1 --- -/--	3,5,25

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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"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

5

Date of the actual completion of the international search	Date of mailing of the international search report
21 April 1994	29. 04. 94

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+ 31-70) 340-2040, Tx. 31 651 epo nl, Fax (+ 31-70) 340-3016	Engel, S

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US,A,5 135 966 (A.M. CHATTERJEE ET AL.) 4 August 1992 see column 2, line 32 - line 34; examples see claims 1,4,8-10	1,3-5, 14,15, 23,25,26
A	US,A,4 156 666 (R. ODATE) 29 May 1979 cited in the application see column 2, line 58 - column 3, line 5 see examples 1,2; table 1 see claims 1,2	1,3-5, 14,15, 23,25,26
A	DE,A,28 40 875 (ASAHI KASEI KOGYO KK) 29 March 1979 see claims 1,4,5	6-10,13
X	see page 7, line 7 - page 10, line 26 see examples; tables	1,3,23
A	WO,A,88 06609 (AMROTEX AG) 7 September 1988 see page 6, line 2 - line 6 see page 12, paragraph 3, lines 23 - 25 see tables 1-3 see claims 1-3	10-12, 20,22
A	EP,A,0 114 724 (ICI) 1 August 1984 see page 1, line 33 - page 2, line 10 see page 6, line 9 - line 13 see page 11, line 21 - line 25; examples see page 15, line 18 - line 20 see claims 1,3,4	10-13

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INTERNATIONAL SEARCH REPORT

International application No.
PCT/US 93/ 07922

Box I Observations where certain claims were found unsearchable (Continuation of item 1 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 II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. Claims 1,14,15,23 (completely); 3-5,25-26 (partially)
2. Cliams 2,16,24 (completely); 3-5,25-26 (partially)
3. Claims 6-13,20-22 (completely)
4. Claims 17-19 (compppletely); 25-26 and 3 (partially)
5. Claims 27,28 (compppletely)
6. Claim 29 (completely)

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 searches without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

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.:

6-13, 20-22

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.

☒ No protest accompanied the payment of additional search fees.

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INTERNATIONAL SEARCH REPORT

Information on patent family members

Inter nal Application No
PCT/US 93/07922

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
US-A-5064893	12-11-91	NONE		
US-A-5135966	04-08-92	NONE		
US-A-4156666	29-05-79	NONE		
DE-A-2840875	29-03-79	JP-C-	1316213	15-05-86
		JP-A-	54047746	14-04-79
		JP-B-	59033138	14-08-84
		US-A-	4219453	26-08-80
WO-A-8806609	07-09-88	CH-A-	671961	13-10-89
		AU-B-	594864	15-03-90
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		DE-A-	3880931	17-06-93
		EP-A,B	0305413	08-03-89
		US-A-	4931488	05-06-90
EP-A-0114724	01-08-84	AU-A-	2329484	19-07-84

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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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A1

(11) International Publication Number: **WO 97/04036**

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(30) Priority Data:
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(81) Designated States: AL, AM, AT, AU, AZ, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, HU, IL, IS, JP, KE, KG, KP, KR, KZ, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, TJ, TM, TR, TT, UA, UG, UZ, VN, ARIPO patent (KE, LS, MW, SD, SZ, UG), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).

Published
With international search report.

(54) Title: NONWOVEN MATERIALS COMPRISING BIODEGRADABLE COPOLYMERS

(57) Abstract

This invention relates to a nonwoven material comprising an adhesive comprising polyhydroxyalkanoate (PHA). This invention also relates to an absorbent article comprising such nonwoven material. This invention also relates to a composition comprising PHA and a surfactant. This invention further relates to a method for preparing the nonwoven.

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NONWOVEN MATERIALS COMPRISING BIODEGRADABLE COPOLYMERS

TECHNICAL FIELD

This invention relates to nonwoven materials and adhesives therefor, comprising biodegradable copolymers.

BACKGROUND

Polymers find uses in a variety of plastic articles including films, sheets, fibers, foams, molded articles, adhesives, nonwoven materials and many other specialty products which usually have a short use cycle. The majority of these products end up in the solid waste stream, headed for limited and increasingly expensive landfill space. While recycling has sought to address this problem, the nature of polymers and polymer products limits the number of possible recycling applications. Repeated processing of polymers and polymer products results in degradation of material and consequently poor mechanical properties. For example, different grades of chemically similar plastics (e.g., polyethylenes of different molecular weights, as used in milk jugs and grocery sacks) mixed upon collection can cause processing problems that make the reclaimed material inferior or unusable.

In absorbent article applications such as diapers, sanitary napkins, pantiliners and the like, the product contains several different types of plastics. In these cases, recycling is particularly costly because of the difficulty in separating the different components. Disposable products of this type generally comprise some sort of fluid-permeable topsheet material, an absorbent core, and a fluid-impermeable backsheet material. Typically, absorbent core materials comprising wood pulp fibers and the like are compostable. However, typical topsheet and backsheet materials prepared from woven, nonwoven, or porous formed-film polyethylene or polypropylene materials are noncompostable. Thus there is a need to replace these noncomposatable materials with compostable material to provide disposable products which are compostable. U.S. Patent No. 4,880,592, Martini et al., discloses a compostable film which, while cumbersome to prepare, is said to be suitable for diaper backsheet applications. An easily prepared, compostable nonwoven, suitable for a diaper topsheet, is desired for compostable product.

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However, any new compostable materials and the articles and products prepared from them must possess certain mechanical properties to facilitate manufacture and consumer acceptance. For example, proper tensile strength, tensile modulus, tear strength, and the like are required for easily preparing the compostable article. The consumer requires properties such as impact strength, puncture strength, and moisture transmission, which influence the compostable article's durability.

Once the absorbent article is disposed of and enters a composting process, other properties become important. The disposable should undergo an initial breakup to much smaller particles during the initial stages of composting.

In the past, the biodegradability and physical properties of a variety of polyhydroxyalkanoates (PHAs) have been studied. PHAs are polyester compounds produced by a variety of microorganisms, such as bacteria and algae. While PHAs have been of general interest because of their biodegradable nature, their use as a plastic material has been hampered by their thermal instability. PHB degrades at temperatures near its melt temperature. Due to this thermal instability, commercial applications of PHB have been limited.

Based on the foregoing, there is a need for nonwoven materials and articles containing them that can biodegrade. To that end, there is a need for a nonwoven material whose fibers are bound together by an adhesive that can biodegrade. In effect, such biodegradable articles would facilitate the "recycling" of plastic articles into another usable product, topsoil, through composting.

Objects of the Invention

It is an object of the present invention to provide a nonwoven material comprising an adhesive comprising a biodegradable polyhydroxyalkanoate (PHA).

It is also an object of the present invention to provide a biodegradable PHA as an adhesive for nonwoven materials.

It is also an object of the present invention to provide a composition comprising a biodegradable PHA useful as an adhesive for nonwoven materials.

It is also an object of the present invention to provide a disposable sanitary article comprising a nonwoven comprising an adhesive comprising a biodegradable PHA.

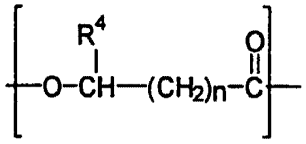
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It is also an object of the present invention to provide a method of using a biodegradable PHA to make nonwoven materials.

SUMMARY

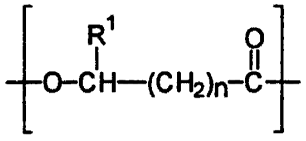
This invention relates to biodegradable PHA copolymers as adhesives useful in preparing nonwovens, and the nonwovens made from them.

The adhesive comprises a PHA of least two different randomly repeating monomer units wherein each randomly repeating monomer unit has the structure:

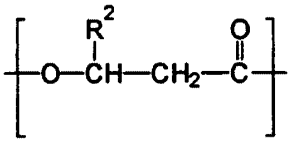


wherein each R^4 is chosen from H, or any of C_1 to about C_{19} alkyl or C_1 to about C_{19} alkenyl and each n is from 1 to about 4.

Preferably, this invention further relates to adhesives useful in preparing nonwovens, and the nonwovens made therefrom, wherein the copolymer comprises at least two randomly repeating monomer units wherein the first monomer unit has the structure;



wherein R^1 is H, or C_1 to about C_{19} alkyl or C_1 to about C_{19} alkenyl, and n is independently 1 or 2; the second monomer unit has the structure:



wherein R^2 is C_3 to about C_{19} alkyl or C_1 to C_{19} alkenyl; and wherein at least 50% of the random repeating monomer units have the structure of the first monomer unit.

More preferably, the first monomer unit has the above structure, wherein R^1 is H, C_1, or C_2 alkyl and n is 1 or 2.

This invention further relates to nonwoven materials and articles comprising them comprising a biodegradable copolymer as an adhesive.

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DETAILED DESCRIPTION

This invention provides a biodegradable nonwoven employing a compostable adhesive as a binder, and disposable articles containing these nonwovens which are easily processed into the article and posses increased biodegradability and/or compostability.

As used herein, "alkyl" means a saturated carbon-containing chain which may be straight or branched; and substituted (mono- or poly-) or unsubstituted. "Substituted," when applied to any radical, refers to any art recognized substitution. Examples of these substitutions are known in the art and can be found in and prepared using, for example, March, Advanced Organic Chemistry, 3rd Ed. (Wiley-Interscience), hereby incorporated by reference, and other similar sources.

As used herein, "unsaturated" means alkyl containing one or more double bonds.

As used herein, "alkenyl" means a carbon-containing chain which may be monounsaturated (i.e., one double bond in the chain) or polyunsaturated (i.e., two or more double bonds in the chain); straight or branched; and substituted (mono- or poly-) or unsubstituted.

As used herein, "PHA" means a polyhydroxyalkanoate, as described throughout the specification.

As used herein, "PHB" means the homopolymer poly-3-hydroxybutyrate.

As used herein, "PHBV" means the copolymer poly(3-hydroxybutyrate-co-3-hydroxyvalerate). This copolymer is also known as PHB-PHV and BIOPOL.

As used herein, "biodegradable" means the ability of a compound to ultimately be degraded completely by microorganisms and/or natural environmental factors.

As used herein, "compostable" means a material that meets the following three requirements:

(1) the material is capable of being processed in a composting facility for solid waste;

(2) if so processed, the material will end up in the final compost; and

(3) if the compost is used in the soil, the material will ultimately biodegrade in the soil.

For example, a polymer material present in solid waste submitted to a composting facility for processing does not necessarily end up in the final compost. Certain composting facilities subject the solid waste stream to air classification prior to further processing, in order to separate paper and other materials. A polymer would most probably be separated from the solid waste stream in such an air classification and therefore not be processed in the composting facility. Nevertheless, it may still be a "compostable" material according to the above definition because it is "capable" of being processed in a composting facility.

The requirement that the material ends up in the final compost typically means that it undergoes a form of degradation in the composting process. Typically, the solid waste stream is subjected to shredding. Here the nonwoven appears in the final compost regardless of composition, biodegradable or not. Therefore, meeting requirement (2) is not enough for a material to be compostable within the present definition.

The compostable material differs from material like polyethylene in that, (3) the material ultimately will biodegrade in the soil. This biodegradability requirement does not require the composting process or the use of composting soil. Nor is it necessary that biodegradation be rapid. Provided that the nonwoven and its decomposition products are non-toxic or polluting, their biodegradation may take months or years.

All copolymer composition ratios recited herein refer to mole ratios, unless specifically indicated otherwise.

As used herein, "plastic article" means a copolymer processed into a film, sheet, fiber, foam, molded article, nonwoven material, elastomer, adhesive, or consumer good, preferably a nonwoven material or consumer good comprising a nonwoven material. A "disposable plastic article" or "disposable article" refers to these articles, when they are likely to be disposable, whether or not they were intended as such. Preferred articles employ the nonwovens of the invention.

As used herein "adhesive" means a material that joins two other materials, called adherents, together. These adherents can be sheets, fibers and the like. Preferably the adherent is a fiber, thus the adhesive binds the fibers together into a nonwoven material.

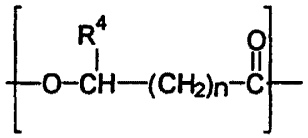
As used herein the term "nonwoven" means, textile-like material, usually in flat sheet form, comprising fibers assembled in webs that are manufactured by processes other than spinning, weaving or knitting. Such

material may be porous or non porous. "Textile like" refers to material used in the manufacture of textiles as well as material made to perform in a manner similar to textiles, without being woven, spun or knitted into textiles. Nonwoven materials include nonwoven fabrics, bonded fabrics, formed fabrics, engineered fabrics, paper-like materials and the like. Nonwoven materials are useful, for example, as topsheets or backsheets for disposable sanitary articles and the like.

As used herein a "disposable sanitary article" includes any object made from the materials described herein that is worn on the body for hygienic or medical purposes. Thus disposable sanitary articles include catamenial products, incontinence products, bandages, wound dressings, surgical pads and the like.

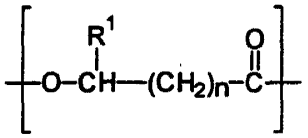
The PHAs disclosed herein are useful as adhesives in nonwovens.

PHAs useful in the present invention comprise at least two randomly repeating monomer units wherein each randomly repeating monomer unit has the structure:

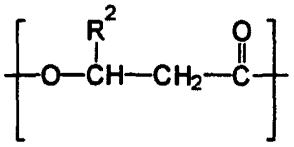


wherein each R^4 is chosen from H, or any of C_1 to about C_{19} alkyl or C_1 to about C_{19} alkenyl and each n is 1 to about 4.

Preferably PHAs useful in the present invention comprise at least two randomly repeating monomer units wherein the first monomer unit has the structure;



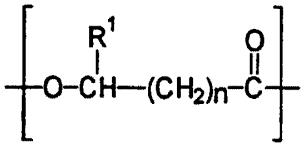
wherein R^1 is H, or C_1 to about C_{19} alkyl or C_1 to about C_{19} alkenyl, and n is independently 1 or 2; the second monomer unit has the structure:



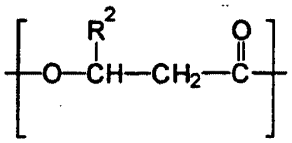
wherein R^2 is about C_3 to about C_{19} alkyl or C_3 to about C_{19} alkenyl; and wherein at least 50% of the random repeating monomer units have the structure of the first monomer unit.

In a preferred embodiment, the first monomer unit has the above structure, wherein R^1 is H, C_1 or C_2 alkyl and n is 1 or 2.

A description of the synthesis of PHAs useful in the present invention and their properties, such as crystallinity, melt temperature and the like, as well as a description of products made using these PHAs is found in copending U.S. Patent Application Serial No. 08/370,738, filed January 12, 1995, incorporated herein by reference. Specifically this application describes PHAs having two randomly repeating monomer units wherein the first randomly repeating monomer unit has the structure

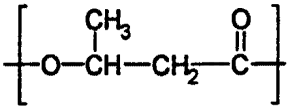


wherein R^1 is H, or C_1 or C_2 alkyl, and n is 1 or 2; the second randomly repeating monomer unit has the structure



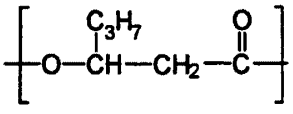
wherein R^2 is a C_3 to about C_{19} alkyl or C_3 to about C_{19} alkenyl; and wherein at least 50% of the randomly repeating monomer units have the structure of the first randomly repeating monomer unit.

Additional PHAs useful in the present invention include those set forth in U.S. Patent Application Serial No. 08/188,271, filed January 28, 1994, incorporated herein by reference. Specifically this application describes PHAs having two randomly repeating monomer units; the first randomly repeating monomer unit having the structure



;

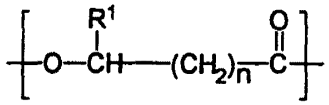
the second randomly repeating monomer unit having the structure



;

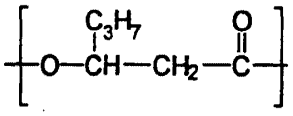
and wherein at least 50% of the randomly repeating monomer units have the structure of the first randomly repeating monomer unit.

Additional PHAs useful in the present invention include those set forth in U.S. Patent Application Serial No. 08/422,011, filed April 13, 1995, incorporated herein by reference. Specifically this application describes PHAs having two randomly repeating monomer units; the first randomly repeating monomer unit having the structure



;

wherein R^1 is H or C_2 alkyl, and n is 1 or 2; the second randomly repeating monomer unit having the structure



and wherein at least 50% of the randomly repeating monomer units have the structure of the first randomly repeating monomer unit. Related disclosure of these PHAs is found in U.S. Patent Application Serial No. 08/188,271, filed January 28, 1994, U.S. Patent Application Serial No. 08/422,011, filed April 13, 1995, U.S. Patent Application Serial No. 08/203,260, filed February 28, 1994, which describe a nonwoven comprising PHA, and method of making it, and U.S. Patent Application Serial No. 08/203,289, filed February 28, 1994, which describes a nonwoven comprising PHA, and method of making it.

The structures of the polymers contained in these references are all within the scope of the structure described in the summary and in the claims.

In one embodiment of the present invention, at least about 50%, but less than 100%, of the randomly repeating monomer units (RRMUs) have the structure of the first RRMU; more preferably at least about 60%; more preferably at least about 70%; more preferably at least about 80%; more preferably still at least about 90%.

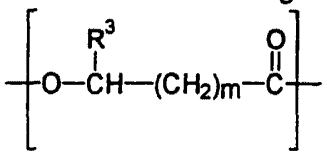
Preferred RRMUs of this embodiment include the following:

R^1	n	Repeating Unit Name
Methyl	1	3-hydroxybutyrate
Ethyl	1	3-hydroxyvalerate
H	2	4-hydroxybutyrate
H	4	6-hydroxycaproate
Methyl	4	6-hydroxycaprylate

H	3	5-hydroxyvalerate
Methyl	3	5-hydroxycaproate

The preceding list of RRMUs for any embodiment is not exhaustive. Certainly other RRMUs are envisioned, all of which are embraced by the claims. Variations of these will be immediately apparent to the skilled artisan.

In another embodiment, the copolymer useful in the present invention comprises one or more additional RRMUs having the structure



wherein R^3 is H, or C_1 to about C_{19} alkyl or C_1 to about C_{19} alkenyl; and m is 1 or 2; and wherein the additional RRMUs are not the same as the first RRMU or the second RRMU. Preferably the copolymer comprises 3 to about 20 or more different RRMUs.

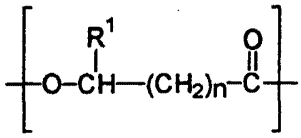
In a preferred embodiment of the present invention, R^3 is a C_1 to about C_{19} alkyl or alkenyl; and m is 1.

Preferred RRMUs of this embodiment include the following:

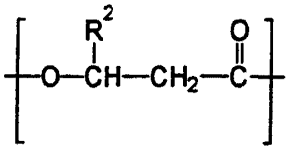
R^3	n	Repeating Unit Name
Methyl	1	3-hydroxybutyrate
Ethyl	1	3-hydroxyvalerate
H	2	4-hydroxybutyrate
H	1	6-hydroxypropionate

The preceding list of RRMUs for any embodiment is not exhaustive. Certainly other RRMUs are envisioned, all of which are embraced by the claims. Variations of these will be immediately apparent to the skilled artisan.

Preferably, the PHAs used in the present invention comprising two RRMUs have a first RRMU having the structure

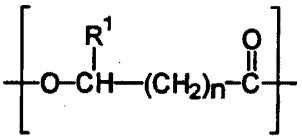


wherein R^1 is H, or C_1 or C_2 alkyl, and n is 1 or 2; and a second RRMU having the structure

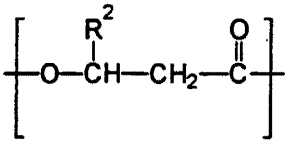


wherein when R^1 is C_1 or C_2 and n is 1, R^2 is a C_{12} to about C_{19} alkyl or alkenyl; when R^1 is C_1 or C_2 and n is 2, R^2 is a C_3 to about C_{19} alkyl or alkenyl; and when R^1 is H and n is 1 or 2, R^2 a C_3 to about C_{19} alkyl or alkenyl; and wherein at least 50% of the RRMUs have the structure of the first RRMU. More preferably R^2 is an alkyl.

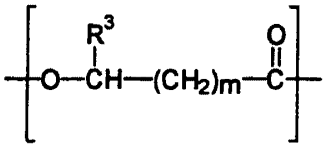
Yet more preferably, novel biodegradable PHAs of the present invention comprising three RRMUs, have a first RRMU having the structure



wherein R^1 is H, or C_1 or C_2 alkyl or C_1 or C_2 alkenyl, and n is independently 1 or 2; a second RRMU having the structure



wherein R^2 is a C_3 to about C_{19} alkyl or C_3 to about C_{19} alkenyl; and a third RRMU having the structure



wherein R^3 is H, or a C_1 to about C_{19} alkyl or C_3 to about C_{19} alkenyl; and m is 1 or 2; wherein at least 50% of the RRMUs have the structure of the first RRMU; and wherein the third RRMU is not the same as the first RRMU or the second RRMU.

Synthesis of biodegradable PHAs, and attendant considerations such as crystallinity, melt temperature and the like are described in U.S. Patent Application Serial No. 08/465,046, filed June 6, 1995, U.S. Patent Application Serial No. 08/422,009, filed April 13, 1995, U.S. Patent Application Serial No. 08/422,008, filed April 13, 1995, U.S. Patent Application Serial No. 08/188,271, filed January 28, 1994, U.S. Patent Application Serial No. 08/422,011, filed April 13, 1995, U.S. Patent

Application Serial No. 08/203,260, filed February 28, 1994 and U.S. Patent Application Serial No. 08/203,289, filed February 28, 1994, all incorporated herein by reference; hereinafter collectively referred to as the "Parent Applications").

The PHAs of the present invention can be processed into a variety of plastic articles, including but not limited to, films, sheets, fibers, foams, molded articles, nonwoven materials, elastomers, and adhesives. Disclosure related to the preparation of such articles is found in the Parent Applications. Of particular interest in this application is the use of PHAs in nonwoven materials preferably their use as adhesives (e.g., binders) in making those nonwoven materials.

Nonwoven Materials and Adhesives Comprising PHA

A. Nonwovens

A general overview of nonwoven materials can be found in the ENCYCLOPEDIA OF POLYMER SCIENCE AND ENGINEERING, Second Edition, Vol. 10, pp. 204-226 (referred to hereafter as "EPSE-4"). The thickness of the nonwoven sheets may vary from 25 μm to several centimeters, and the weight from 10 g/m^2 to 1 kg/m^2.

In contrast to conventional textiles, the fundamental structure of all nonwovens is a web of fibers arranged more or less randomly (NONWOVENS IND., Vol. 17, p. 36 (Mar. 1986), NONWOVENS WORLD, Vol. 1, p. 36 (May-June 1986)). Nonwoven materials have a wide range of physical properties depending on the material and process used in forming the fiber web foundation of the nonwoven. This material may be self-supporting and stiff like paper or drapable like a conventional cloth. Tensile, tear, and tactile properties in the nonwoven arise from the fiber and adhesive or other chemical and physical bonding, fiber-to-fiber friction created by entanglement, and reinforcement by other materials such as foams and films (see EPSE-4).

1. Method of Manufacture of Nonwoven Materials

The nonwoven materials of the present invention may be made by conventional techniques known in the art. Typically the production of nonwoven materials involves: 1) making fibers of various lengths and diameters; 2) creating a web of these fibers; and 3) bonding of fibers within the web by adhesive (sometimes called a binder), or by mechanical-frictional forces created by fiber contact or entanglement, although this process need not necessarily take place in this order. In addition to these

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steps, reinforcing the web by forming a composite with other materials (e.g., yarns, scrims, films, nettings, and unbonded webs) is sometimes preferred. Variations of one or several of these steps or their order allows for the enormous range of processes and of nonwoven fiber types.

The term "staple fibers" was originally applied to fibers of natural origin long enough to be processed on textile machinery, but excluding endless filaments, e.g., silk. In the present context, "staple fibers" are of relatively uniform length, ca. 1.3-10.2 cm, with a regular crimp i.e., a three-dimensional wavelike shape. Regenerated and other extruded fibers which are endless as formed, are typically cut during the manufacturing process to meet a processing or market need or specification. Extruded fibers can also be produced as continuous filaments without crimp.

The processes for forming webs from staple fibers differ from those using continuous filaments. Nonwoven material obtained from staple and filament fiber webs may differ substantially in properties (see EPSE-4).

The mechanical properties of the fibers, as well as of the adhesives used to adhere the nonwoven fibers to each other influences the properties of the nonwoven (see EPSE-4).

Other fibrous materials that may be used in the nonwovens of the present invention in combination with the PHA are natural fibers such as silk, wool, linen and the like: cellulosics, such as wood pulp, paper fibers, cotton, cellulose and the like; regenerated fibers including viscose rayon and cellulose acetate and the like; and synthetic fibers like polyesters, such as poly(ethylene terephthalate) (PET), other PHAs and the like; the polyamides, such as nylon-6, nylon 6,6, and the like; polyalkylenes, such as polypropylene (PP), polyethylene (PE) and other synthetics such as poly(vinyl alcohol) and the like.

The choice of fiber depends upon the intended application. For example, facings of disposable diapers or sanitary napkins made from PHA nonwoven materials of the present invention preferably feel dry even when the absorbent, inner layer is saturated. Hence, a hydrophobic polymer, such as a polyester, polyamide, polyalkylene and the like, is preferred for use in these products.

For applications where water absorbency is desirable, hydrophilic fibers are preferred. For example, a cellulosic fiber or a polyvinyl alcohol fiber or the like would be preferred in making a nonwoven towelette.

For applications where fluid impermeability is desirable, the nonporous nonwoven preferably employ:

1) fibers which repel fluids;

2) a web dense enough to provide aviod holes or pores in the nonwoven;

3) enough adhesive to seals any holes or pores in the nonwoven web,

Such nonporous nonwoven materials are useful for mailing envelopes, containers, vapor barriers, diaper backsheets and the like

Other important fiber characteristics that affect performance include length, diameter, density, crimp, cross section shape, spin-finish (e.g., lubricant added to the surface of extruded fibers to enhance processability), delustering (e.g., small amounts of TiO_2 pigment added before extrusion to increase whiteness or to reduce sheen) and the draw ratio. These are all related to the fiber or the fiber web, and are discussed in EPSE-4.

These nonwovens can be made by several methods, for example the methods of Noda et al. exemplified in U.S. patent applications 08/203,260, filed February 28, 1994, and 08/203,289, filed February 28, 1994, incorporated herein by reference, as well as art recognized methods.

a. Web-making methods

The characteristics of the fiber web influence the physical properties of the final product. These characteristics depend on fiber architecture, which is determined by the mode of web formation. Fiber architecture includes the predominant fiber direction, whether oriented or random, fiber shape (straight, hooked, or curled), the extent of interfiber engagement or entanglement, crimp, and compaction (web-density control). Web characteristics are also influenced by fiber diameter, length, web weight, and chemical and mechanical properties of the fiber material (see EPSE-4).

The choice of method for forming the web is determined primarily by fiber length. Initially, the methods for forming webs from staple-length fibers (fibers long enough to be handled by conventional spinning equipment, usually from about 1.2 to about 20 cm long, but not endless) are based on the textile-carding process, whereas web formation from short fibers is based on papermaking technologies. Although these technologies are still in use, other methods have been subsequently developed. For example, webs are formed from long, virtually endless filaments directly from bulk polymer; both web and fibers are produced simultaneously (see EPSE-4). A

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variety of web-making methods are known, including carding, air-laying, wet-forming, spunbonding, and meltblowing.

Carding separates clumps of staple fibers mechanically into individual fibers and formed into a coherent web by the mechanical action of moving beds of closely spaced needles.

Air-laying improves orienting of fibers by capturing fibers on a screen from an airstream (see U.S. Patent No. 3,338,992, G. A. Kinney, assigned to E.I. du Pont de Nemours & Co., Inc., issued August 29, 1967). The fibers are separated by teeth or needles and introduced into an airstream.

Wet-forming typically employs very short fibers. Initially, webs are formed from short fibers by modified papermaking techniques. The fibers are continuously dispersed in a large volume of water or solvent and caught on a moving endless wire screen. Once the web is caught on the screen, it is transferred to belts or felts and dried on heated drums (see EPSE-4).

Spunbonding involves making fibers and web simultaneously, directly from bulk polymer. The bulk polymer is melted, extruded, and drawn into virtually endless filaments that are randomized and deposited onto belts as a continuous web. This process produces webs of low crimp filaments in the normal diameter range of about 1.7 dtex (1.5 den) or slightly higher. The birefringence and uniformity of diameter of these filaments are similar to standard textile fibers and filaments (see EPSE-4 and U.S. Pat. 4,163,305 (Aug. 7, 1979), V. Semjonow and J. Foedrowitz (to Hoechst AG)).

Meltblowing prepares webs directly from bulk polymers (see U.S. Patent No. 3,322,607, S.L. Jung, assigned to E.I. duPont de Nemours & Co., Inc., May 30, 1967). For example, molten PHA forced through very fine holes in a special die into a high velocity airstream forms very fine, although irregular, filaments of indeterminate lengths. The filaments are formed into a web and bonded simultaneously (see EPSE-4). The web consists primarily of very fine filaments.

b. Web bonding

The bonding of fibers (i.e., the adhering of fibers to each other) gives the strength to the web and influences the nonwoven properties. Both adhesive and mechanical means are used in bonding. Mechanical bonding uses bonding of fibers by frictional forces. Bonding can also be achieved by chemical means, e.g., formation of covalent bonds between binder and fibers (see EPSE-4).

In a preferred method, this bonding is accomplished with a compostable material, more preferably a PHA, as an adhesive. Such a preferred method of bonding of fibers used in a nonwoven material is described herein below.

B. Adhesive

A general discussion on adhesives can be found in the Encyclopedia of Polymer Science and Engineering, Vol. 1, pp. 547-577, (hereafter referred to as "EPSE-6"). Generally an adhesive is applied as a liquid, preferably of a low viscosity. The adhesive wets the adherent surface and may flow into the crevices in the adherent surfaces. In many cases, the liquid form of the adhesive is obtained by heating the adhesive to the point that flow occurs, dissolving or dispersing the material in a solvent, or starting with liquid monomers or oligomers that polymerize or react after application. Usually, the adhesive then solidifies by cooling, solvent evaporation, reaction or the like to provide the necessary strength to resist shearing forces.

The PHAs of the present invention may be processed into a variety of adhesives, including but not limited to, hot melt, solution, dispersion and pressure sensitive adhesives.

The method of using these adhesives and the choice of adhesive will depend upon the properties sought, the material to which the adhesive is applied and the application intended. For example, in preparing a nonwoven material, one would consider the mechanical characteristics of the unbonded nonwoven and its desired end application.

1. Hot-melt Adhesives.

As used herein, "hot-melt adhesive" refers to a thermoplastic polymer or copolymer (e.g., a PHA) that is heated to obtain a liquid of flowable viscosity, and, after application, cooled to obtain a solid. Generally, the molecular weight of the adhesive is tailored to provide good rheology as a melt and sufficient strength as a solid to resist shearing forces experienced in the application. The PHAs of the present invention are thermoplastic, and thus are particularly useful as hot-melt adhesives. The primary feature of hot-melt adhesives is the ability of the thermoplastic material (PHA) to flow above a certain temperature, and to provide a strong bond at the normal use temperature. Upon cooling, the material hardens, either through passing through the glass transition temperature or the crystallization temperature. This hardening lends physical integrity to the bond.

2. Solutions and Dispersions.

The adhesives of this invention may be applied either as solutions or in the form of aqueous dispersions. When using a solution or dispersion, heating is preferred to expedite drying or "setting" of the adhesive. The dispersion solids can vary from 5 to 95%, preferably less than 50%.

As used herein, "dispersion" refers to adhesives which are prepared or dispersed as larger particles in some carrier fluid. In addition to their economic advantage, dispersions containing 40-50% solids offer lower viscosity than solutions, even if the solids are high molecular-weight polymers (EPSE-6). Adhesive dispersions of the present invention may be dispersed by high shear in the presence of surfactants to obtain waterborne formulations by procedures which are well known to those skilled in the art.

One preferred method of using the adhesive is to prepare a solution or dispersion of the PHA in toluene, ethyl acetate, acetone, halogenated solvents (e.g., chloroform, 1,2 dichloroethane and the like) or other suitable solvent. To this solution or dispersion it is preferred that a surfactant be added. Surfactants may increase wetting ability of the adhesive, as well as dispersion characteristics. Preferably the surfactant is an anionic surfactant, cationic surfactant, zwitterionic surfactant, nonionic surfactant, amphoteric surfactant or the like, preferably a cationic surfactant. Preferred surfactants are described in U.S. Patent No's. 4,835,211, Noda et al., issued May 30, 1989, and 4,785,030, Noda et al., issued November 15, 1988, incorporated herein by reference.

A more preferred surfactant is a surfactant of formula X-Y-Z; wherein X is a cationic radical chosen from primary, secondary or tertiary amines and their cations, quaternary ammonium, sulfonium and phosphonium and the like; Y is a nonionic diradical having up to about 20 repeating monomer units of $-(OCH_2CH_2)-$; and Z is an alkyl or alkenyl radical comprising of from about 8 to about 20 carbons which is branched or linear and substituted or unsubstituted.

In a preferred embodiment, this surfactant is present in about 0.5% to about 20% by weight; preferably about 1% to about 10% by weight; more preferably about 2% to about 5% by weight. Where a surfactant is used, water may be added to the solution or dispersion. Where water is added, it may be preferable to remove any solvent in the solution or dispersion. This provides an "environmentally friendly" adhesive.

3. Pressure-sensitive Adhesives.

These adhesives are discussed in detail in U.S. Patent application Serial No. 08/465,046, filed June 6, 1995, incorporated herein by reference.

4. Use of adhesives as binders in nonwovens

PHAs can be used as adhesives or "binders" in nonwoven materials. For longer fiber nonwovens, it is preferred that the nonbonded material be formed and then coated using the art recognized "dip" coating process, wherein the nonwoven is run through a bath of adhesive in a dispersion or solution, preferably a colloidal dispersion in water. The adhesive then dries or sets and is optionally pressed to provide the bonded nonwoven.

For materials lacking the mechanical integrity to withstand dip coating, it is preferred that spray coating be used. Spray coating uses a sprayed adhesive, preferably a heated water dispersion of the adhesive, which then dries or cures and is optionally pressed to provide the bonded nonwoven.

For materials which are made from fibers that are too short to be spray coated, the fibers themselves can be coated in a bath and the nonwoven formed from these adhesive coated materials. In this case, the nonwoven material typically cannot be formed in the absence of binder. Spray coating would likely disperse the fibers. Consequently, coating the fibers in a bath is preferred, for example, in making paper like materials.

In a preferred embodiment of the present invention, the adhesive is dispersed in an aqueous medium with the aid of a surfactant. Preferably, the surfactant will promote the spontaneous deposition of the adhesive to fibers. In a more preferred embodiment, the surfactant is of formula X-Y-Z; wherein X is a cationic radical chosen from primary, secondary or tertiary amines and their cations, quaternary ammonium, sulfonium and phosphonium and the like; Y is a nonionic diradical having up to about 20 repeating monomer units of $-(OCH_2CH_2)-$, preferably 5 to about 20, more preferably about 10 to about 20; and Z is an alkyl or alkenyl radical comprising of from about 8 to about 20 carbons which is saturated or

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unsaturated, branched or linear, preferably saturated or monounsaturated, and of from about 15 to about 20 carbons.

Disposable Personal Care Products

The nonwovens described above are useful in many disposable products: For example, disposable sanitary articles comprising a liquid pervious topsheet comprising the nonwoven comprising the PHA of the invention, a liquid impervious backsheet, and an absorbent core positioned between the topsheet and backsheet. Such articles include infant diapers, adult incontinent briefs and pads, and feminine hygiene pads, liners and the like.

Additional personal care products comprising a PHA of the present invention include personal cleansing wipes; disposable health care products such as bandages, wound dressings, wound cleansing pads, surgical gowns, surgical covers, surgical pads; other institutional and health care disposables such as gowns, wipes, pads, bedding items such as sheets, pillowcases, and foam mattress pads.

Additional absorbent articles include paper products, such as towelettes, tissues, paper towels, disposable washcloths and the like: personal care products, such as sanitary napkins, diapers, incontinence briefs and the like; health care products, such as wound dressings, bandages, and the like. Many other absorbent articles are known in the art and are contemplated.

Importantly, the absorbent articles of this invention are biodegradable and/or compostable to a greater extent than conventional absorbent articles which employ standard materials, such as polyolefins, and the like.

The following examples are not intended to limit or define the invention in any way. The following nonlimiting examples merely illustrate the invention and teach the skilled artisan how to make and use the invention. Of course, it is obvious to the skilled artisan that variation of these examples, including any permutation claimed in this application can be prepared by the skilled artisan in light of the examples following.

Additional examples of preparation of PHAs used herein below, as well as further disclosure and examples of the manufacture of these articles and examples thereof, including the preparation of films, sheets, fibers, foams, molded articles and elastomers are described in the Parent Applications.

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EXAMPLE 1

Compostable Nonwoven Fabric

Poly(3-hydroxybutyrate-co-3-hydroxyoctanoate) (PHB-O) at 2 mole% octanoate/ 98 mole% butyrate is introduced into a single screw extruder (Rheomix Model 202, Paramus, NJ) with screw diameter of 0.75 inch. A constant taper screw having 20:1 length to diameter ratio and a 3:1 compression ratio is employed. The temperature of both heating zones of the extruder barrel is 25°C above the melt temperature of the PHB-O. The extruder is equipped with a nozzle die containing 5 orifices of diameter 500 mm. The die is maintained at 20°C above the melt temperature of the PHB-O. The polymer is melted within the extruder and pumped to the die at the other end of the extruder. The screw rpm is kept constant at 30 rpm. The polymer is forced through the die and the melted extruded fibers are lead through a region where a rapid air stream is applied such that the polymer fibers elongates and thins to approximately one fifth of the diameter of the orifices (ca. 100 μm). The fibers are collected on a cardboard mat. The mat is moved in a fashion so that a 10 cm x 10 cm area is covered uniformly with fibers. Collection of fibers on the mat continues, until there is approximately 0.5 cm thick fiber mat. A wide distribution of fiber lengths are obtained up several inches in length. Most fiber lengths (over 50%) are in the range of 0.5 to 6 inches. The mat is then transferred to a Carver Press (Fred S. Carver Inc., Menomonee Falls, WI) and pressed at a 1000 lb. force for 10 minutes at temperature 5°C below the melting temperature of the PHB-O. The resulting nonwoven sheet is removed from the press.

EXAMPLE 2

Compostable Adhesive

PHB-O (50:50) may be used as a hot-melt adhesive in the following manner. About 1g of PHB-O (50:50) is placed between two polymer films, such as poly(vinyl alcohol) (PVA), or poly(3-hydroxybutyrate) (PHB) or any other PHA which has a melting temperature at least 10°C higher than PHB-O (50:50). The films/adhesive assembly is placed in a Carver Press (Fred S. Carver Inc., Menomonee Falls, WI) and is then pressed at a temperature 5°C above the melt temperature of PHB-O (50:50). After compression at 2000 lb. force for 30 min., the pressure is released and the bonded film assembly is allowed to cool to room temperature.

EXAMPLE 3

A. Dispersion of PHA Binder

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A solution made of 50 g of 92:8 poly(3-hydroxybutyrate-co-hydroxyoctanoate) copolymer dissolved in 450 g of an organic solvent such as chloroform is emulsified in 2 liters of water with the aid of sonicator. An emulsifier, e.g., oleyl ethoxylate as VOLPO-20 (Croda, Inc.) can be used in the mixture to stabilize the emulsion. The organic solvent is subsequently evaporated from the emulsion to obtain a dispersion of the copolymer binder.

B. Nonwovens Made by Dip Coating of PHA Binder

2.62 g (2.5 g dry wt.) unrefined Northern Softwood Kraft (NSK) pulp is dispersed in 500 ml tap water at ambient pH (ca. 7.5). The handsheet is made on a standard Deckle Box using tap water at ambient pH (ca. 7.5) and dried on a drum drier at 115°C. The handsheet is then immersed in the dispersion of PHA binder as described above and dried again to produce a nonwoven sheet.

EXAMPLE 4

Compostable Nonwoven Fabric

Using the PHB-O of Example 1 and the hot melt adhesive of Example 2, a compostable nonwoven fabric is prepared by placing the fibers collected on a cardboard mat with the adhesive into a Carver Press as in Example 1, thus forming a nonwoven wherein the fiber and the binder is of PHA.

EXAMPLE 5

A. Dispersion of Cationic PHA Binder

A solution made of 50 g of 98:2 poly(3-hydroxybutyrate-co-hydroxyoctadecanoate) copolymer dissolved in 450 g of an organic solvent such as chloroform is emulsified in 2 liters of water with the aid of sonicator. An emulsifier, e.g., oleyl ethoxylate as VOLPO-20 can be used in the mixture to stabilize the emulsion. To the emulsion 1.0 g of a cationic emulsifier, e.g. oleyl ethoxylate end-capped with a quaternary ammonium moiety as described in patents granted to Noda et al. (U.S. Pat. No's. 4,835,211, issued May 30, 1989, and 4,785,030, issued November 15, 1988) is added. The organic solvent is subsequently evaporated from the emulsion to obtain a cationic dispersion of the copolymer binder.

B. Nonwoven Fabrics Made by Wet-End Deposition of PHA Binder

2.62 g (2.5 g dry wt.) unrefined Northern Softwood Kraft (NSK) pulp is dispersed in 500 ml tap water at ambient pH (ca. 7.5). 1.0 g of cationic binder dispersion described in A is added to the pulp slurry and stirred for 30 minutes to promote the deposition. The handsheet is made on a standard Deckle Box using tap water at ambient pH (ca. 7.5) and dried on a drum drier at 115°C.

EXAMPLE 6

A. Dispersion of Cationic PHA Binder

A solution made of 50 g of 90:10 poly(3-hydroxybutyrate-co-hydroxyhexanoate) copolymer dissolved in 450 g of an organic solvent such as chloroform is emulsified in 2 liters of water with the aid of sonicator. An emulsifier, e.g., oleyl ethoxylate as VOLPO-20 can be used in the mixture to stabilize the emulsion. To the emulsion 1.0 g of a cationic emulsifier, e.g. oleyl ethoxylate end-capped with a quaternary ammonium moiety as described in patents granted to Noda et al. (U.S. Pat. No's. 4,835,211, issued May 30, 1989, and 4,785,030, issued November 15, 1988) is added. The organic solvent is subsequently evaporated from the emulsion to obtain a cationic dispersion of the copolymer binder.

B. Nonwoven Fabrics Made by Wet-End Deposition of PHA Binder

2.62 g (2.5 g dry wt.) unrefined Northern Softwood Kraft (NSK) pulp is dispersed in 500 ml tap water at ambient pH (ca. 7.5). 1.0 g of cationic binder dispersion described in A is added to the pulp slurry and stirred for 30 minutes to promote the deposition. The handsheet is made on a

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standard Deckle Box using tap water at ambient pH (ca. 7.5) and dried on a drum drier at 115°C.

EXAMPLE 7

A. Dispersion of Cationic PHA Binder

A solution made of 50 g of 92:8 poly(3-hydroxybutyrate-co-hydroxyoctanoate) copolymer dissolved in 450 g of an organic solvent such as chloroform is emulsified in 2 liters of water with the aid of sonicator. An emulsifier, e.g., oleyl ethoxylate as VOLPO-20 can be used in the mixture to stabilize the emulsion. To the emulsion 1.0 g of a cationic emulsifier, e.g. oleyl ethoxylate end-capped with a quaternary ammonium moiety as described in patents granted to Noda et al. (U.S. Pat. No's. 4,835,211, issued May 30, 1989, and 4,785,030, issued November 15, 1988) is added. The organic solvent is subsequently evaporated from the emulsion to obtain a cationic dispersion of the copolymer binder.

B. Nonwovens Made by Wet-End Deposition of PHA Binder

2.62 g (2.5 g dry wt.) unrefined Northern Softwood Kraft (NSK) pulp is dispersed in 500 ml tap water at ambient pH (ca. 7.5). 1.0 g of cationic binder dispersion described in A is added to the pulp slurry and stirred for 30 minutes to promote the deposition. The handsheet is made on a standard Deckle Box using tap water at ambient pH (ca. 7.5) and dried on a drum drier at 115°C.

EXAMPLE 8

Continuous Production of Nonwovens with PHA Binder

The applicability of cationic dispersion of PHA as a binder for nonwovens produced by wet-end deposition process is as follows. Approximately 220 kg (dry weight) of refined northern Kraft pulp is dispersed in water at the consistency of about 2.5% and kept in a stirred holding tank. About 400 liters of cationic binder dispersion (prepared according to Example 7A) is added to the pulp to achieve the wet-end deposition of the binder.

The binder-treated pulp is then fed to a pilot-scale paper machine (equipped with normal papermaking process components, such as headbox, forming wire, and continuous dryer) at a rate of 80L/min. The paper machine is operated at the production speed of 200 m/min.

EXAMPLE 9

Compostable Disposable Diaper

A disposable baby diaper according to this invention is prepared as follows. The dimensions listed are for a diaper intended for use with a child in the 6-10 kilogram size range. These dimensions can be modified proportionately for different size children, or for adult incontinence briefs, according to standard practice.

1. Backsheet: 0.020 - 0.038 mm film consisting of a 92:8 poly(3-hydroxybutyrate-co-hydroxyoctanoate) copolymer; width at top and bottom 33 cm; notched inwardly on both sides to a width-at-center of 28.5 cm; length 50.2 cm.

2. Topsheet: a topsheet of nonwoven fabric of Example 8: width at top and bottom 33 cm; notched inwardly on both sides to a width-at-center of 28.5 cm; length 50.2 cm.

3. Absorbent core: comprises 28.6 g of cellulose wood pulp and 4.9 g of absorbent gelling material particles (commercial polyacrylate from Nippon Shokubai); 8.4 mm thick, calendered; width at top and bottom 28.6 cm; notched inwardly at both sides to a width-at-center of 10.2 cm; length 44.5 cm.

4. Elastic leg bands: four individual rubber strips (2 per side); width 4.77 mm; length 370 mm; thickness 0.178 mm (all the foregoing dimensions being in the relaxed state).

The diaper is prepared in standard fashion by positioning the core material covered with the topsheet on the backsheet and gluing.

The elastic bands (designated "inner" and "outer", corresponding to the bands closest to, and farthest from, the core, respectively) are stretched to ca. 50.2 cm and positioned between the topsheet/backsheet along each longitudinal side (2 bands per side) of the core. The inner bands along each side are positioned ca. 55 mm from the narrowest width of the core (measured from the inner edge of the elastic bank). This provides a spacing element along each side of the diaper comprising the flexible topsheet/backsheet material between the inner elastic and the curved edge of the core. The inner bands are glued down along their length in the stretched state. The outer bands are positioned ca. 13 mm from the inner bands, and are glued down along their length in the stretched state. The topsheet/backsheet assembly is flexible, and the glued-down bands contract to elasticize the sides of the diaper.

EXAMPLE 10

Compostable Lightweight Pantiliner

A lightweight pantiliner suitable for use between menstrual periods comprises a pad (surface area 117 cm^2; SSK air felt 3.0 g) containing 1.0 g of absorbent gelling material particles (commercial polyacrylate; Nippon Shokubai); said pad being interposed between a porous topsheet of Example 8 according to U.S. Patent 4,463,045 and a backsheet which comprises a 0.03 mm thickness 92:8 poly(3-hydroxybutyrate-co-hydroxyoctanoate) copolymer film.

EXAMPLE 11

Compostable Sanitary Napkin

A catamenial product in the form of a sanitary napkin having two flaps extending outward from its absorbent core is prepared using a pad in the manner of Example 10 (surface area 117 cm^2; 8.5 g SSK air felt), per the design of U.S. Patent 4,687,478, Van Tillburg, August 18, 1987.

EXAMPLE 12

Compostable Disposable Diaper

The diaper of Example 9 is modified by replacing the backsheet with a backsheet consisting of a 0.020 to 0.038 mm thickness film comprising a 92:8 poly(3-hydroxybutyrate-co-hydroxydecanoate) copolymer film.

EXAMPLE 13

Compostable Disposable Diaper

The diaper of Example 12 is modified by replacing the nonwoven backsheet with a backsheet made from the nonwoven of Example 4.

EXAMPLE 14

Compostable Disposable Diaper

The diaper of Example 12 is modified by replacing the nonwoven topsheet with a nonwoven porous topsheet prepared by the methods disclosed in copending U.S. Patent application 08/203,260, incorprated herein by reference.

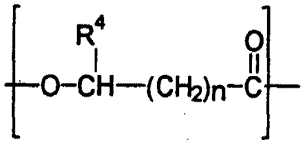
All publications and patent applications mentioned hereinabove are hereby incorporated in their entirety by reference.

It is understood that the examples and embodiments described herein are for illustrative purposes only and that various modifications or changes in light thereof will be suggested to one skilled in the art and are to be included in the spirit and purview of this application and scope of the appended claims.

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WHAT IS CLAIMED IS:

1. An adhesive dispersion comprising a biodegradable copolymer characterized in that the biodegradable copolymer comprises at least two randomly repeating monomer units characterized in that each randomly repeating monomer unit has the structure:



wherein R^4 is H, or C_1 to C_{19} alkyl or C_1 to C_{19} alkenyl, and n is 1 to 4.

2. The adhesive dispersion of any preceding claim, further characterized by a surfactant of formula:

X-Y-Z

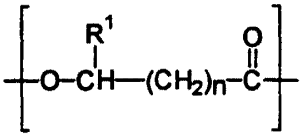
wherein;

X is a cationic radical chosen from the group consisting of primary, secondary or tertiary amines and their cations, and quaternary ammonium, and sulfonium and phosphonium;

Y is a nonionic diradical having up to 20 repeating monomer units of -(OCH_2CH_2)-;

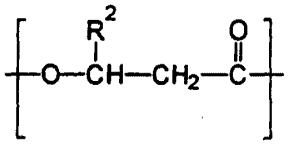
Z is an alkyl or alkenyl radical comprising of from 8 to 20 carbons which is branched or linear and substituted or unsubstituted.

3. The adhesive dispersion of any preceding claim, where the biodegradable polymer is further characterized by the two randomly repeating monomer units wherein a first randomly repeating monomer unit has the structure:



wherein R^1 is H, or C_1 to C_{19} alkyl or C_1 to C_{19} alkenyl, and n is 1 or 2; and a second randomly repeating monomer unit having the structure:

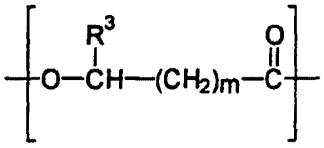
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wherein R^2 is C_3 to C_{19} alkyl or C_3 to C_{19} alkenyl; and wherein at least 50% of the randomly repeating monomer units have the structure of the first randomly repeating monomer unit.

4. The adhesive dispersion of any preceding claim, wherein R^1 is H, or C_1 or C_2 alkyl, and n is 1 or 2, and R^2 is C_3 to C_{19} alkyl or C_3 to C_{19} alkenyl.

5. The adhesive dispersion of any preceding claim, wherein the biodegradable copolymer further comprises one or more additional randomly repeating monomer units having the structure:



wherein R^3 is H, or a C_1-C_{19} alkyl or C_1 to C_{19} alkenyl; and m is 1 or 2; and wherein the additional randomly repeating monomer units are not the same as the first randomly repeating monomer unit or the second randomly repeating monomer unit.

6. A nonwoven material further comprising fibers; and the adhesive of any preceding claim.

7. A liquid pervious topsheet comprising the nonwoven material of claim 6.

8. An absorbent article comprising the adhesive dispersion or nonwoven material of any preceding claim.

9. The absorbent article of claim 8 for personal hygiene.

10. A method of making the nonwoven material of Claim 1, comprising the steps of:

a) combining the biodegradable polymer with a surfactant of formula:

X-Y-Z

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wherein;

X is a cationic radical chosen from the group consisting of primary, secondary or tertiary amines and their cations, and quaternary ammonium, and sulfonium and phosphonium;

Y is a nonionic diradical having up to 20 repeating monomer units of

$-(OCH_2CH_2)-$;

Z is an alkyl or alkenyl radical comprising of from 8 to 20 carbons which is saturated or unsaturated, branched or linear and substituted or unsubstituted;

b) coating a fiber with the binder; and

c) bonding the fibers together into a nonwoven material.

INTERNATIONAL SEARCH REPORT

Interr ial Application No
PCT/US 96/11864

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C09J167/04 D04H1/64

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 6 C09J C09D D04H

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 practical, 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	TAPPI JOURNAL, vol. 76, no. 5, May 1993, ATLANTA US, pages 71-77, XP000369471 C. A. LAUZIER ET AL.: "Film formation and paper coating with poly(beta-hydroxyalkanoate), a biodegradable latex" see page 75, middle column, line 3 - line 10 see page 71, left-hand column, line 1 - middle column, line 2 ---	1,2,4, 6-10
X	DE,A,40 40 158 (PCD PETROCHEMIE DANUBIA DEUTSCHLAND GMBH) 17 June 1992 see claims 1-4; examples 5,21,35 ---	1,2,4, 6-10
A	WO,A,95 02649 (ZENECA) 26 January 1995 see claims 1-11 -----	1-10

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in 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 document 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
5 November 1996	14. 11. 96
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+ 31-70) 340-2040, Tx. 31 651 epo nl, Fax (+ 31-70) 340-3016	Authorized officer Decocker, L

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INTERNATIONAL SEARCH REPORT

Information on patent family members

Inter nal Application No
PCT/US 96/11864

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
DE-A-4040158	17-06-92	NONE		
WO-A-9502649	26-01-95	AU-A-	7129294	13-02-95
		CA-A-	2166613	26-01-95
		EP-A-	0708804	01-05-96
		FI-A-	960158	12-01-96
		NO-A-	960153	12-01-96

Form PCT/ISA/210 (patent family annex) (July 1992)

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APPLICATION NUMBER	FILING OR 371(c) DATE	FIRST NAMED APPLICANT	ATTY. DOCKET NO./TITLE
11/348,150	02/06/2006	Christopher S. Cleveland	FDS-029388-US

CONFIRMATION NO. 5681

1726
INTERNATIONAL PAPER COMPANY
6285 TRI-RIDGE BOULEVARD
LOVELAND, OH45140

Title: Biodegradable paper-based laminate with oxygen and moisture barrier properties and method for making biodegradable paper-based laminate

Publication No. US-2007-0184220-A1
Publication Date: 08/09/2007

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 Office of Public Records. The Office of Public Records can be reached by telephone at (703) 308-9726 or (800) 972-6382, by facsimile at (703) 305-8759, by mail addressed to the United States Patent and Trademark Office, Office of Public Records, 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 http://pair.uspto.gov/. 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.

Pre-Grant Publication Division, 703-605-4283

JRW

INTERNATIONAL PAPER

THOMAS W. BARNES, Ph.D
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INTELLECTUAL PROPERTY



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October 23, 2007

Commissioner for Patents
Post Office Box 1450
Alexandria, Virginia 22313-1450

RE: SUBMISSION OF INFORMATION DISCLOSURE STATEMENT

Applicant	:	CLEVELAND, et al.
Serial No.	:	11/348,150
Filed on	:	FEBRUARY 6, 2006
Title	:	BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOISTURE.....
Our Ref.	:	FDS-029388-US

Dear Commissioner:

Enclosed herewith for filing, Applicant(s) respectfully submit(s) the following checked items:

☐ Information Disclosure Statement by Applicant(s), _____ sheet(s), to which Form PTO/SB/08, _____ sheet(s), has been attached along with a copy of each reference(s) cited thereon;

☒ Supplemental Information Disclosure Statement by Applicant(s), 4 sheet(s), to which Form PTO/SB/08, 1 sheet(s), has been attached along with a copy of each reference(s) cited thereon; and,

☒ a postage-prepaid, return-receipt postcard with a listing of the above-checked items.

Please stamp the enclosed postcard and return same to me to indicate your receipt of the above-listed items. Please feel free to contact me if you have any questions concerning the above or the enclosed.

With kindest regards,

Thomas W. Barnes
Agent for Applicant(s)

Enclosures
TWB/ jat

CERTIFICATE OF MAILING OR TRANSMISSION UNDER 37 C.F.R. § 1.8

The undersigned hereby certifies that a true and accurate copy of the documents checked hereon, together with all attachments referred-to herein, is being transmitted to the Honorable Commissioner for Patents, either by first-class mail, postage prepaid, addressed to Commissioner for Patents, Post Office Box 1450, Alexandria, Virginia 22313-1450, or by facsimile transmission to the facsimile number indicated hereon, on this the 23rd day of October 20 07

JANE A. TOMLINSON

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
WASHINGTON, D.C., UNITED STATES OF AMERICA

In re Application of:

Applicant(s): CLEVELAND, et al.

Serial No.: 11/348,150

Filing Date: FEBRUARY 6, 2006



Entitled: BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOSITURE .

Examiner:

Our File Ref.: FDS-029388-US

Group Art Unit: 17721

SUPPLEMENTAL INFORMATION DISCLOSURE STATEMENT
SUBMITTED PURSUANT TO 37 C.F.R. 1.97(b)(3)

Commissioner for Patents
United States Patent and Trademark Office
Post Office Box 1450
Alexandria, Virginia 22313-1450

Dear Commissioner:

In accordance with the duty of disclosure under 37 C.F.R. 1.56, Applicant(s) hereby notifies the U.S. Patent and Trademark Office of certain documents which the Examiner might deem relevant to the issue of patentability of the claims pending in the above-referenced patent application. Enclosed herewith please find Form PTO/SB/08, Information Disclosure Statement by Applicant, 4 sheet(s), and a copy of each reference cited thereon.

This Information Disclosure Statement is filed before the mailing date of a first Office Action on the merits. As such, neither a fee under 37 C.F.R. 1.17(p) nor a statement under 37 C.F.R. 1.97(e) is required; Applicant(s) respectfully requests pursuant to 37 C.F.R. 1.97(b)(3) that the Examiner considers each of the references disclosed herewith.

The submission of the documents enclosed herewith is not intended as an admission that such documents constitute prior art under Title 35 of the United States Code, as against the claims of the above-referenced patent application. Applicant(s) does not waive any right to take

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action that would be appropriate to antedate or otherwise overcome the use of any document submitted herewith as prior art under Title 35 of the United States Code.

Applicant(s) respectfully requests that the Examiner initial each of the references listed on the attached Form PTO/SB/08 and return same to the undersigned attorney in accordance with MPEP 609.

Should the Examiner have any questions relating to the submission of the within Information Disclosure Statement or relating to the copies submitted therewith, Applicant(s) respectfully request that the Examiner contact the undersigned counsel, who can be reached at the telephone numbers listed below.

Respectfully submitted,

Date: 10/23/2007

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 1 of 4

Complete if Known	
Application Number	11/348,150
Filing Date	FEBRUARY 6, 2006
First Named Inventor	CLEVELAND, et al.
Art Unit	1772
Examiner Name	
Attorney Docket Number	FDS-029388-US

U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] *(if known)*	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
	A	US- 3,901,838	08-26-1975	Clendinning, et al	
	B	US- 5,096,941	03-17-1992	Robert M. Harnden	
	C	US- 5,416,133	05-16-1995	Garcia, et al	
	D	US- 5,458,933	10-17-1995	Stuart P. Suskind	
	E	US- 5,498,385	03-12-1996	Yabusa, et al	
	F	US- 5,534,616	07-09-1996	Simon D. Waddington	
	G	US- 5,565,503	10-15-1996	Garcia et al	
	H	US- 5,593,778	01-14-1997	Kondo, et al	
	I	US- 5,602,227	02-11-1997	Isao Noda	
	J	US- 5,618,855	04-08-1997	Isao Noda	
	K	US- 5,620,281	04-15-1997	Lammers, et al	
	L	US- 5,685,756	11-11-1997	Isao Noda	
	M	US- 5,693,389	12-02-1997	John J. Liggat	
	N	US- 5,854,304	12-29-1998	Garcia, et al	
	O	US- 5,939,467	08-17-1999	Wnuk, et al	
	P	US- 6,033,747	03-07-2000	Takeshi Shiotani	
	Q	US- 6,040,063	03-21-2000	Doane, et al	
	R	US- 6,080,478	06-27-2000	Hannu Karhuketo	
	S	US- 6,096,810	08-01-2000	Asrar, et al	

FOREIGN PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Foreign Patent Document Country Code[3]-Number[4]-Kind Code[5] *(if known)*	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages Or Relevant Figures Appear	T[6]
	F1	JP2002-322307	11-08-2002			
		English				
	F2	JP2003-276144	09-30-2003			
		English				
	F3	JP2003-41142	02-13-2003			
		English				

Examiner Signature		Date Considered	

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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 2 hours 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, 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.**

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Sheet 2 of 4

Complete if Known	
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Art Unit	1772
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Attorney Docket Number	FDS-029388-US

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	T	US- 6,111,006	08-29-2000	Simon D. Waddington	
	U	US- 6,140,422	10-31-2000	Khanarian, et al	
	V	US- 6,235,815	05-22-2001	Loercks, et al	
	W	US- 6,245,437	06-12-2001	Shiiki, et al	
	X	US- 6,248,862	06-19-2001	Asrar, et al	
	Y	US- 6,323,010	11-27-2001	Skraly, et al	
	Z	US- 6,326,440	12-04-2001	Terada, et al	
	A1	US- 6,342,304	01-29-2002	Buchanan, et al	
	A2	US- 6,414,108	07-02-2002	Warzelhan, et al	
	A3	US- 6,458,858	10-01-2002	Braun, et al	
	A4	US- 6,521,336	02-18-2003	Narita, et al	
	A5	US- 6,569,990	05-27-2003	Isao Noda	
	A6	US- 6,573,340	06-03-2003	Khemani, et al	
	A7	US- 6,586,354	07-01-2003	Topolkaraev, et al	
	A8	US- 6,600,008	07-29-2003	Kobayashi, et al	
	A9	US- 6,610,764	08-26-2003	Martin, et al	
	B1	US- 6,617,415	09-09-2003	Miksic, et al.	
	B2	US- 6,620,869	09-16-2003	Asrar, et al	
	B3	US- 6,730,057	05-04-2004	Zhao, et al	

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Examiner Signature		Date Considered	

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Sheet 3 of 4

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Attorney Docket Number	FDS-029388-US

U. S. PATENT DOCUMENTS

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	B4	US- 2001/0003761	06-14-2001	Ishibashi, et al	
	B5	US- 2003/0040695	02-27-2003	Zhao, et al	
	B6	US- WO 92/01733	02-06-1992	Webb, et al	
	B7	US- WO 94/04606	03-03-1994	Garcia, et al	
	B8	US- WO 97/04036	02-06-1997	Isao Noda	
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Sheet 4 of 4

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	A	US- WO/06/031568	03-23-2006	Cleveland, et al	
	B	US- WO/01/61043	08-23-2001	Jian-Bing Fan	
	C	US- WO/04/082936	09-30-2004	Rieghard, et al	
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	F1	EP 0629975	12-21-1994			
	F2	DE 19954404	11-12-1999			
		WO 01/36518				
	F3	JP 04299131	10-22-1992			
		English				

Examiner Signature		Date Considered	

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(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2002－322307
（P2002－322307A）
(43)公開日　平成14年11月８日(2002.11.8)

(51)Int.Cl.⁷		識別記号	FI			ﾃｰﾏｺｰﾄﾞ(参考)
C08J	9/12	CEP	C08J	9/12	CEP	3E066
B65D	65/46		B65D	65/46		3E067
	81/107			81/28	C	3E086
	81/28			85/34	G	3E096
	85/34		C08J	9/36		4F074

審査請求　未請求　請求項の数２　書面　（全　４　頁）　最終頁に続く

(21)出願番号　特願2001－163457(P2001－163457)
(22)出願日　平成13年４月23日(2001.4.23)

(71)出願人　596035097
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(74)代理人　100085914
弁理士　竹田　逸郎

最終頁に続く

(54)【発明の名称】　包装用生分解性樹脂発泡体

(57)【要約】
【課題】果物等を包装するため使用されている発泡ポリスチレンシートに代替しうる包装用生分解性樹脂発泡シートを開発し、廃棄物問題の解決に寄与する。
【解決手段】主として、セルロースアセテート３５～９０重量%、生分解性コポリエステル５～６０重量%、非木材質植物繊維粉末２～２０重量%、高温処理した竹の幹の繊維粉末２～２０重量%、及び、低重合度ポリエチレングリコール１～１０重量%からなることを特徴とする包装用生分解性樹脂発泡体、ならびに、その表面に天然の抗菌剤を塗布したことを特徴とする包装用生分解性樹脂発泡体を提供する。

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【特許請求の範囲】
【請求項１】　主として、セルロースアセテート３５～９０重量%、生分解性コポリエステル５～６０重量%、非木材質植物繊維粉末２～２０重量%、高温処理した竹の幹の繊維粉末２～２０重量%、及び、低重合度ポリエチレングリコール１～１０重量%からなることを特徴とする包装用生分解性樹脂発泡体
【請求項２】　請求項１記載の包装用生分解性樹脂発泡体の表面に、更に天然の抗菌剤を塗布したことを特徴とする包装用生分解性樹脂発泡体
【発明の詳細な説明】
【０００１】
【発明の属する技術分野】本発明は、主として、梨、りんご、洋梨等の果物を包むために使用される発泡ポリスチレンシートや、それを収納するプラスチックケースや、ダンボール箱の内装に使用される緩衝シート等に代替しうる包装用生分解樹性脂発泡体に関するものである。
【０００２】
【従来の技術】従来、梨、りんご、洋梨等の果物は、和紙や籾のついた紙で包んで木綿等の緩衝材が充填された箱の中に入れて輸送されてきたが、最近になって、これらの果物は、薄手の発泡ポリスチレンシートで包んでから、プラスチックケースに詰められ、それをダンボール箱に入れて、上から発泡ポリスチレンシート等で覆い、蓋をしてダンボール箱に収納して輸送することが多くなった。
【０００３】
【発明が解決しようとする課題】前述のように、果物等を包むために薄手の発泡ポリスチレンシートや、プラスチックケースや、緩衝用の発泡ポリスチレンシートは、使用後に廃棄し処理するに当たって、嵩高のため、収集し運搬するにも、また、投棄場に投棄するにも大きな容積を占め、また、焼却するにしても高温になり炉を損傷する等の問題があった。そのため、多種多様な生分解性樹脂や生崩壊性樹脂が開発されてきた。しかしながら、これらの生分解樹脂又は生崩壊樹脂は、ポリスチレンの異なり、予備発泡させた後、型に入れて本発泡させて成型品を作ることが困難であった。そのため多くの技術が開発されてきた。
【０００４】生分解性樹脂発泡体の成型品を製造する方法には、小発泡体を接着させて成型品を製造する方法と成型品の形状に発泡させる方法がある。小発泡体を接着させる方法には、本出願人が特開平１０－３３０５２７号公報で開示した方法や特開２０００－１４１５００号公報で開示した方法等があり、所定の形状に発泡させる方法には、本出願人が特願平２０００－３０２９１０号公報で開示した方法等がある。この公報に開示した方法は、押出機のダイに隣接して設けられた所定の断面形状の外周に内接する成型枠の中で発泡させるものであり、包装用に使用されるシート状の生分解樹脂発泡成型品も製造することができる。しかしながら、得られたシート状の製品は、長手方向には高い引張強度を有しているが、横方向には、長手方向の引張強度よりかなり低く、引き裂け易いという欠点があった。なお、この問題を解決するため、フィルムを製造するように、Ｔダイから樹脂を押し出して発泡させて発泡生分解樹脂シートを製造したところ、確かに、横方向の引張強度が高くなったが、必ずしも充分なものではなかった。
【０００５】このような問題を解決するため、本発明者は、特願２０００－２４３４１９号明細書で開示したように、セルロースアセテートに、引張強度を上げるため非木材質繊維を混入し、更に、柔軟性を上げるためポリエチレングリコールを添加して、それなりの引張強度を有するシート状の生分解性樹脂発泡体を製造することができた。しかしながら、果物等の包装用シートとしては、必ずしも、引張強度や柔軟性の点で充分なものではなかった。本発明が解決しようとする課題は、更に引張強度や柔軟性を高め、果物の鮮度保持性も付加して、果物等を包んだり、果物等のケースにしたり、果物等の緩衝材としたりすることができる包装用の生分解性樹脂発泡成型体を提供することにある。
【０００６】
【課題を解決するための手段】本発明者は、前述の課題を解決するために、主として、セルロースアセテート３５～９０重量%、生分解性コポリエステル５～６０重量%、非木材質植物繊維粉末２～２０重量%、高温処理した竹の幹の繊維粉末２～２０重量%、及び、低重合度ポリエチレングリコール１～１０重量%からなることを特徴とする包装用生分解性樹脂発泡体（以下「第１発明」という）、ならびに、第１発明に係わる包装用生分解性樹脂発泡体の表面に、更に天然の抗菌剤を塗布したことを特徴とする包装用生分解性樹脂発泡体（以下「第２発明」という）を提供する。
【０００７】第１発明におけるセルロースアセテートは、セルロースの水酸基を無水酢酸等でアセチル化したもので、酢化の程度によって各種のものがある。このセルロースアセテートは、発泡体の骨格をなすものであり、このような目的に使用できるものであれば、どのようなセルロースアセテートでもよい。通常は、市販のセルロースアセテートが、そのまま使用される。生分解性コポリエステルは、脂肪鎖と芳香環を有するポリエステルであって生分解性を有するものある。この生分解性コポリエステルは、骨格となるセルロースアセテートの補強剤として作用し、非木材質植物繊維粉末、及び、高温処理した竹の幹の繊維粉末の接着剤として作用するものと考えられる。現在、イーストマンケミカル社の「イースターバイオ」やＢＡＳＦ社の「エコフレックス」等が知られている。また、生分解性コポリエステルの中には、「イースターバイオ」のように、１００℃以上の高

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い融点を有するものもある。

【０００８】このセルロースアセテートと生分解性コポリエステルとの比率は、セルロースアセテートが３５～９０重量%、生分解性コポリエステルが５～６０重量%の範囲であり、生分解性コポリエステルが、これより少ないと、補強効果や接着効果が現れず、また、これより多くしても、これれらの効果がそれに比例して増強されなくなる。また、コストも高くなる。

【０００９】また、非木材質植物繊維粉末は、草本類をパルプ化したものであり、（株）チャフローズコーポレーション製「チャフローズ」が知られている。この非木材質植物繊維粉末を添加することにより、横方向の引張強度と柔軟性とを高めることが期待される。高温処理した竹の幹の繊維粉末は、皮を剥いだ竹の幹を高温処理して粉末化した繊維粉末（以下「竹鮮」という）であり、（株）タケックス製「パデオール」が知られている。この竹鮮を添加することにより、横方向の弾力性と引張強度を高め、果物等の被包装物の鮮度保持性を持たせることが期待される。

【００１０】これらの非木材質植物繊維粉末及び竹鮮の添加比率は、非木材質植物繊維粉末が２～２０重量%、竹鮮が２～２０重量%の範囲であり、これらの範囲未満であれば、縦方向及び横方向の引張強度を高める効果が現れず、また、これらの範囲を超えても、それに比例して引張強度が高くならなくなる。また、シート状に成型するのも困難になる。

【００１１】また、低重合度ポリエチレングリコールは、医薬品や化粧品に使用されているものであり、通常ポリエチレングリコール４００が用いられる。この低重合度ポリエチレングリコールは発泡体に弾性や柔軟性を与えるものである。また、その添加比率は１～１０重量%の範囲で充分である。その他に添加されるものには、ほたて貝殻を高熱処理して得られた抗菌性、天然色素、薬草粉末、薬草を煎じた液、天然の顔料等があげられる。

【００１２】第２発明に係わる包装用生分解性樹脂発泡体は、第１発明に係わる包装用生分解性樹脂発泡体の表面に、更に天然の抗菌剤を塗布したことを特徴としている。この天然の抗菌剤には、穀物から得られた天然フラボノイド系抗菌剤として、ニッセキ（株）製「セントリスＥ－１」等が知られている。

【００１３】第１発明に係わる包装用生分解樹脂発泡体を製造するには、主として、セルロースアセテート３５～９０重量%に水を添加して膨潤させ、非木材質植物繊維粉末２～２０重量%、竹鮮２～２０重量%、低重合度ポリエチレングリコール１～１０重量%、及び、生分解性コポリエステル１５～３０重量%を添加して、エクストルーダーに入れ、加熱溶融してＴダイから押し出し、断面が幅の広い矩形状になっている筒状の成形枠の中で発泡させて、シート状の成形品にする方法がとられる。

【００１４】第２発明に係わる包装用生分解性樹脂発泡体を製造するには、前述の方法で得られたシート状の成型品の両面又は片面に、天然の抗菌剤の水溶液を噴霧する等の方法により、天然の抗菌剤を塗布する方法がとられる。

【００１５】

【発明の実施の態様】１．第１発明に係わる包装用生分解樹性脂発泡体等の製造

〔実施例１〕セルロースアセテート（帝人（株）製「アセテートフレーク」）６．７ｋｇに水０．７ｋｇを添加して膨潤させ、非木材質植物繊維粉末（（株）チャフローズコーポレーション製「チャフローズ」）０．３ｋｇ、竹鮮（（株）タケックス製「パデオール」）０．５ｋｇ、ポリエチレングリコール（４００）０．５ｋｇ、及び、生分解性コポリエステル（イーストマンケミカル社製「イースターバイオＧＰ」）２．０ｋｇを加えて混合し、二軸押出機（φ４６ｍｍ、Ｌ／Ｄ２１．５）に供給し、シリンダー温度$C_1$８５℃、$C_2$１２４℃、$C_3$１３１℃、$C_4$１４０℃、$C_5$１９４℃、$C_6$１８１℃、$D_1$１７１℃、$D_2$１８０℃で、幅１００ｃｍ、スリット間隔１ｍｍのノズルから押し出し、発泡させて、幅１００ｃｍ、厚さ２．０ｍｍの第１発明に係わる包装用生分解性樹脂発泡体（以下「シートＡ」という）を製造した。

【００１６】〔比較例〕セルロースアセテート（帝人（株）製「アセテートフレーク」）７．８ｋｇに水０．７ｋｇを添加して膨潤させ、非木材質植物繊維粉末（（株）チャフローズコーポレーション製「チャフローズ」）０．８ｋｇ、および、ポリエチレングリコール（４００）２．０ｋｇとを加えて混合し、二軸押出機（φ４６ｍｍ、Ｌ／Ｄ２１．５）に供給し、シリンダー温度$C_1$７０℃、$C_2$９０℃、$C_3$１３０℃、$C_4$２００℃、$C_5$２１０℃、Ｄ１９０℃で、幅１００ｃｍ、スリット間隔１ｍｍのノズルから押し出し、発泡させて、幅１００ｃｍ、厚さ２．０ｍｍの比較のための包装用生分解性樹脂発泡体（以下「シートＢ」という）を製造した。

【００１７】２．第２発明に係わる包装用生分解樹性脂発泡体の製造

〔実施例２〕実施例１で得られたシートＡ」の両面に、天然フラボノイド系抗菌剤（ニッセキ（株）製「セントリスＥ－１」）２．０重量%水溶液を噴霧して、第２発明に係わる包装用生分解樹性脂発泡体（以下「シートＣ」という）を製造した。

【００１８】３．引張強度等の特性の比較

実施例１で得られたシートＡは、比較例で得られたシートＢに比べて、ポリエチレングリコールの添加量が少ないにもかかわらず、表面が滑らかで、柔軟性があった。また、シートＡとシートＢとのそれぞれについて、１０個の縦１２０ｍｍ、横１２０ｍｍ（厚さ２．０ｍｍ）の

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サンプルを作成して、それぞれ、その中の５個について、縦方向の一端を固定し他端をばね秤を介して引っ張り、切断した時のばね秤の表示を縦方向の引張強度として測定した。また、それぞれ残り５個について、横方向の一端を固定して、同様にして横方向の引張強度を測定した。その結果、縦方向の平均の引張強度は、従来品のシートＢは３．５ｋｇであるのに対して、本発明に係わるシートＡは５．５ｋｇに向上していた。また、横方向の平均の引張強度は、従来品のシートＢは１．８ｋｇであるのに対して、本発明に係わるシートＡは２．０ｋｇに向上していた。

【００１９】このように、縦方向の引張強度も横方向の引張強度が向上したのは、生分解性コポリエステルが、添加された非木材質植物繊維粉末や高温処理した竹の幹の繊維粉末を、セルロースアセテート発泡体中に接着する作用を果たしているためと考えられる。また、縦方向の引張強度が大きく向上するのに対して、横方向の引張強度の向上が僅かであるのは、添加された非木材質植物繊維粉末や高温処理した竹の幹の繊維粉末が、Ｔダイから押し出される際に流れ方向に配向する傾向にあって、それが発泡の際にもあまり乱されないためと考えられる。

【００２０】４．鮮度保持試験

実施例２で得られた第２発明に係わるシートＣ１０枚、及び、従来品のシートＢ１０枚で、それぞれ、１０個の林檎を包み、１週間室温で保管したところ、従来品のシートＢで包んだ林檎には黄味を帯びていたが、シートＣで包んだ林檎には全く変化が見られなかった。

【００２１】

【発明の効果】本発明に係わる包装用生分解性樹脂発泡体は、前述のような構成であって、包装材料に適した引張強度を有し、かつ、果物等に対する鮮度保持作用を有するので、これまで果物の包装に使用されてきた発泡ポリスチレンシートやプラスチックケースに代替して、果物等の包装等に幅広く使用することができ、プラスチック廃棄物に関する問題の解決に寄与し、環境の保全に貢献し、国民の生活の向上に大きく貢献するものである。

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(71)Applicant : PLUSTO:KK
(72)Inventor : HAYASHIBARA HITOSHI

(54) BIODEGRADABLE RESIN FOAM FOR PACKAGING

(57)Abstract:
PROBLEM TO BE SOLVED: To contribute to solve waste problems by developing a biodegradable resin foam sheet for packaging capable of replacing a formed polystyrene sheet used for packaging fruits, or the like.
SOLUTION: This biodegradable resin foam mainly comprises 35-90 wt.% of a cellulose acetate, 5-60 wt.% of a biodegradable co-polyester, 2-20 wt.% of a non-wood plant fiber powder, 2-20 wt.% of a bamboo-trunk fiber powder treated at a high temperature and 1-10 wt.% of a low polymerization degree polyethylene glycol. The biodegradable resin foam for packaging applied with a natural anti-bacterial agent on the surface is also provided.

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最終頁に続く

(54)【発明の名称】　生分解性ポリエステル積層フィルム

(57)【要約】
【課題】生分解性を損なうことなく、防湿性が改良された生分解性ポリエステル積層フィルムを提供する。
【解決手段】ポリ乳酸等の脂肪族ポリエステルからなる生分解性ポリエステルフィルム基材層の少なくとも片面に好ましくは粘度平均分子量が１０００～７０００の範囲にある低分子量ポリオレフィン層が積層されてなる、生分解性を損なうことなく、防湿性が改良された生分解性ポリエステル積層フィルムに関する。

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【特許請求の範囲】
【請求項１】生分解性ポリエステルフィルムからなる基材層の少なくとも片面に低分子量ポリオレフィン層が積層されてなることを特徴とする生分解性ポリエステル積層フィルム。
【請求項２】生分解性ポリエステルフィルムが、二軸延伸されてなる請求項１記載の生分解性ポリエステル積層フィルム。
【請求項３】低分子量ポリオレフィンが、粘度平均分子量が１０００～７０００の範囲にある請求項１記載の生分解性ポリエステル積層フィルム。
【請求項４】低分子量ポリオレフィンが、低分子量エチレン系重合体である請求項１又は３記載の生分解性ポリエステル積層フィルム。
【請求項５】生分解性ポリエステルが、ポリ乳酸である請求項１記載の生分解性ポリエステル積層フィルム。
【請求項６】生分解性ポリエステルが、芳香族系ポリエステルである請求項１記載の生分解性ポリエステル積層フィルム。
【請求項７】芳香族系ポリエステルがスルホン酸金属塩基を核置換基として有する芳香族ジカルボン酸を含む芳香族系ポリエステルである請求項６記載の生分解性ポリエステル積層フィルム。
【発明の詳細な説明】
【０００１】
【産業上の利用分野】本発明は、防湿性が改良された生分解性ポリエステル積層フィルムに関する。
【０００２】
【従来の技術】生分解性フィルムは、土壌中や水中で加水分解や生分解を受け、徐々に崩壊や分解が進み、最後には微生物の作用で分解物へと変化するものである。そのようなフィルムとして、ポリ乳酸等の脂肪族系ポリエステル樹脂や芳香族系ポリエステル樹脂、ポリビニルアルコール、酢酸セルロース、デンプン等から成形したフィルムが知られており、中でも、ポリ乳酸を用いた二軸延伸フィルムは、包装用フィルムとして大量に用いられているニ軸延伸ポリプロピレンフィルムに似て、透明性、剛性に優れることから、包装用フィルムとして一部で使用されている。しかしながら、かかるポリ乳酸二軸延伸フィルムは、ニ軸延伸ポリプロピレンフィルムに比べると防湿性（水蒸気バリヤー性）に劣ることから包装用フィルムとしての利用範囲が限られている。
【０００３】生分解性ポリエステルフィルムの防湿性を改良する方法として、ポリ乳酸を主成分とするフィルムと再生セルロースフィルムとからなるフィルムが提案されている（特開平５－３８７８４号公報）が、再生セルロース自体がポリオレフィン等に比べ防湿性に劣ることから、得られるフィルムは未だ防湿性が不十分である。
【０００４】
【発明が解決しようとする課題】そこで本発明は、ポリ乳酸等の脂肪族系ポリエステル樹脂や芳香族系ポリエステル樹脂等の生分解性ポリエステルフィルムが本来有する生分解性を損なうことなく、防湿性に優れた二軸延伸フィルムを開発することを目的とした。
【０００５】
【課題を解決するための手段】
【発明の概要】本発明は、生分解性ポリエステルフィルムからなる基材層の少なくとも片面に低分子量ポリオレフィン層が積層されてなることを特徴とする防湿性に優れた生分解性ポリエステル積層フィルムを提供することにある。そして、好ましくは、生分解性ポリエステルとして、ポリ乳酸及び芳香族ポリエステル、特にスルホン酸金属塩基を核置換基として有する芳香族ジカルボン酸を含む芳香族系ポリエステルから選択され、低分子量ポリオレフィンとして粘度平均分子量が１０００～７０００の範囲にあり、又、低分子量ポリオレフィンが低分子量エチレン系重合体である生分解性ポリエステル積層フィルムを提供することにある。
【０００６】
【発明の具体的な説明】生分解性ポリエステル
本発明の基材層を構成する生分解性ポリエステルフィルムの原料である生分解性ポリエステルは、種々公知の生分解性ポリエステルであり、具体的にはポリ乳酸、あるいはコハク酸、グルタル酸、アジピン酸、ピメリン酸、スベリン酸、アゼライン酸、セバシン酸、ウンデカン二酸、ドデカン二酸、シクロペンタンジカルボン酸およびシクロヘキサンジカルボン酸等の通常、炭素数が４～１２の二塩基性カルボン酸とエチレングリコール、トリエチレングリコール、１，４－ブタンジオール、ペンタメチレングリコール、１，８－オクチレングリコール、ネオペンチレングリコール、デカメチレングリコール等の通常、炭素数が２～１０のグリコールから任意に選ばれた１種あるいは２種以上のジカルボン酸とグリコールの脱水縮合あるいはそれに続く脱グリコール反応により得られる生分解性を有する脂肪族ポリエステル、あるいは芳香族ジカルボン酸、脂肪族グリコール、およびスルホン酸金属塩基を核置換基として有する芳香族ジカルボン酸からなる生分解性を有する芳香族系ポリエステルである。これらの中でも、融点が１５０℃以上のポリ乳酸及び芳香族ポリエステル、特にスルホン酸金属塩基を核置換基として有する芳香族ジカルボン酸を含む芳香族系ポリエステルが、防湿性能、酸素バリア性能、透明性、強度、柔軟性といったフィルム物性に優れるので好ましい。
【０００７】ポリ乳酸
生分解性ポリエステルの例である本発明に係わるポリ乳酸は、乳酸の構造単位がＬ－乳酸であるポリ（Ｌ－乳酸）、構造単位がＤ－乳酸であるポリ（Ｄ－乳酸）、更にはＬ－乳酸とＤ－乳酸の混合物（ラセミ体）であるポリ（ＤＬ－乳酸）等の乳酸の単独重合体、及び乳酸を主

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成分とし、乳酸以外の共重合可能なコモノマーを少割合、例えばグリコール酸５０重量%未満、好ましくは３０重量%以下、より好ましくは１０重量%以下の割合で共重合したコポリマー、あるいはこれらの混合物等の乳酸を主成分とする重合体である。乳酸と共重合可能なコモノマーとしては、例えば３－ヒドロキシブチレート、カプロラクトン、グリコール酸などを挙げることができる。これらの中でも乳酸のみからなる単独重合体は透明性に優れているので好ましく使用できる。かかるポリ乳酸の重合法としては、縮重合法、開環重合法など公知のいずれの方法を採用することができる。例えば、縮重合法ではＬ－乳酸またはＤ－乳酸あるいはこれらの混合物を直接脱水縮重合して任意の組成を持ったポリ乳酸を得ることができる。

【０００８】また、開環重合法では乳酸の環状２量体であるラクチドを、必要に応じて重合調整剤等を用いながら、触媒を使用してポリ乳酸を得ることができる。ラクチドにはＬ－乳酸の２量体であるＬ－ラクチド、Ｄ－乳酸の２量体であるＤ－ラクチド、さらにＬ－乳酸とＤ－乳酸からなるＤＬ－ラクチドがあり、これらを必要に応じて混合して重合することにより任意の組成、結晶性を有するポリ乳酸を得ることができる。

【０００９】ポリ乳酸の分子量増大を目的として少量の鎖延長剤、例えば、ジイソシアネート化合物、エポキシ化合物、酸無水物などを使用できる。ポリ乳酸の重量平均分子量は通常６万～１００万の範囲にあり、この範囲を下回る場合は用途によっては実用物性がほとんど発現されない虞があり、一方、上回る場合には、溶融粘度が高すぎ成形加工性に劣る虞がある。

【００１０】芳香族ポリエステル

生分解性ポリエステルの他の例である本発明に係わる芳香族ポリエステルは、生分解性を有するものであり、好ましくはスルホン酸金属塩基を核置換基として有する芳香族ジカルボン酸を１共重合成分として含むポリエステルであり、より具体的には、芳香族ジカルボン酸、脂肪族グリコール、およびスルホン酸金属塩基を核置換基として有する芳香族ジカルボン酸、それに必要に応じて脂肪族ジカルボン酸または脂肪族ヒドロキシカルボン酸を加え、それらの成分間で重縮合反応を行って得られるポリエステルであることが望ましい。芳香族ポリエステルの好ましい組成は、芳香族ジカルボン酸成分に由来する単位が３０～４９．９モル%、脂肪族グリコール成分に由来する単位が３５～５０モル%、スルホン酸金属塩基を置換基として有する芳香族または脂肪族ジカルボン酸成分に由来する単位が０．１～５モル%、および脂肪族ジカルボン酸または脂肪族ヒドロキシカルボン酸成分に由来する単位が０～３０モル%（ここで、全単位の合計が１００モル%になる）である。また得られるフィルムに可撓性、生分解性等の性能を付与し、また向上させるために、さらに脂肪族ジカルボン酸または脂肪族ヒドロキシカルボン酸を共重合成分として加えた多成分系のポリエステルであってもよい。そのような芳香族ポリエステルは、特表平５－５０７１０９号公報、特表平６－５０５０４０号公報、特表平６－５０５５１３号公報等に詳しい。

【００１１】本発明に係わる芳香族ポリエステルは、その重量平均分子量が、１０，０００～５００，０００の範囲が好ましい。また、そのメルトフローレートは、ＡＳＴＭ Ｄ－１２３８に準拠し、２２０℃、２１６０ｇ荷重下で測定した値が、０．１～１００（ｇ／１０分）であることが好ましい。分子量およびメルトフローレートが前記の範囲内にあると、押出成形に適した溶融粘度を示し、また二軸延伸フィルムとしての十分な機械的強度を有する。

【００１２】低分子量ポリオレフィン

本発明の生分解性ポリエステルフィルムからなる基材層の少なくとも片面に積層される低分子量ポリオレフィン層となる低分子量ポリオレフィンは、エチレン、プロピレン、１－ブテン、１－ヘキセン、１－オクテン、４－メチル－１ペンテン等の炭素数２から１０のα－オレフィンの単独若しくは共重合体であり、通常、粘度平均分子量が１０００～７０００、好ましくは１０００～６０００の範囲にある。粘度平均分子量が１０００未満のものは、基材層に積層して得られる積層フィルムの耐摩耗性、塗膜強度等に劣る虞があり、一方、７０００を越えるものは、得られる積層フィルムの生分解性が低下する傾向にある。また、これら低分子量ポリオレフィンは、種々公知の方法、例えば、溶融状態にあるワックスを、約１２０乃至２００℃、好ましくは約１４０乃至１７０℃で酸素含有ガスと接触させ、酸化した酸化物であっても、あるいは、低分子量ポリオレフィンにマレイン酸、無水マレイン酸等の不飽和カルボン酸またはその誘導体成分をグラフト共重合してなるグラフト変性物であってもよい。

【００１３】これら、低分子量オレフィン重合体の中でも、エチレンの単独重合体あるいはエチレンを主体とした重合体等のエチレン系重合体が好ましく、特に融点が９０～１３５℃、より好ましくは１１０～１３５℃の範囲にあり、且つ、密度が０．９０～０．９５ｇ／ｃｍ^3、より好ましくは０．９１～０．９４ｇ／ｃｍ^{3}の範囲にあるものが防湿性に優れる。またかかる低分子量ポリオレフィンには得られる積層フィルムの防湿性能をより向上させる目的で粒径０．１～５μｍのヘクトライト等の無機層状化合物を配合してもよい。また、得られる積層フィルムの生分解速度を向上させる目的で他の各種生分解性のプラスチックを配合してもよい。

【００１４】生分解性ポリエステル積層フィルム

本発明の生分解性ポリエステル積層フィルムは、生分解性ポリエステルフィルムからなる基材層の少なくとも片面に低分子量ポリオレフィン層が積層されてなる。基材

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層及び低分子量ポリオレフィン層の厚さは用途に応じて種々決定されるが、通常、基材層の厚さが、２０～１００μｍ、好ましくは２５～５０μｍ、低分子量ポリオレフィン層の厚さが、０．１～５μｍ、好ましくは０．５～３μｍの範囲にある。

【００１５】本発明の生分解性ポリエステル積層フィルムは種々公知の方法で得られるが、低分子量ポリオレフィンは溶融粘度が低いので、通常は、予め製造した生分解性ポリエステルフィルムの少なくとも片面に、低分子量ポリオレフィンの水性分散体を被覆して乾燥する方法が好ましい。基材層となる生分解性ポリエステルフィルムは無延伸フィルムでも、一軸あるいは二軸延伸フィルムでもよいが、二軸延伸フィルムが得られる積層フィルムの機械的強度、防湿性等に優れるので好ましい。

【００１６】低分子量ポリオレフィンの水性分散体としては、通常、粒子径が０．０１～１０μｍ、好ましくは０．１～９μｍの低分子量ポリオレフィン粒子を水に分散した分散体である。又、基材層に被覆（塗布）した後、乾燥を速やかに行えるようにアルコールを０～５０溶液重量%、好ましくは１０～３０溶液重量%含む水性分散体にしておいても良い。低分子量ポリオレフォンの水性分散体には生分解性ポリエステルフィルムとの接着性若しくは濡れ性改良のために水系イソシアネート系接着材、セロソルブ系等の有機溶剤、粘度調整剤、防かび剤、静防剤などを添加することができる。水性分散体の濃度は、コーティングに支障のない範囲で高い方が良い。通常１０～５０%、好ましくは２０～４０%である。最適な粘度は塗布方法や製造速度により異なるが、例えば、グラビアロールコーティングにおいては２００ｃｐｓ以下が好ましい。

【００１７】低分子量ポリオレフィン水性分散体の塗布方法は、従来使用されているものが利用できる。例えば、ダイレクトグラビアコーティング、グラビアオフセットコーティング、アークグラビアコーティング、グラビアリバースコーティング等のグラビアコーティング、トップフィードリバースロールコーティング、ボトムフィードリバースロールコーティング、ノズルフィードリバースロールコーティング等のリバースロールコーティング、キスロールコーテイング、マイヤーバーコーティング、ドクターブレードコーティング、及びエアーナイフコーティングなどが挙げられる。これらの中でもグラビアコーティング、特にグラビアリバースコーティングが好ましい。

【００１８】生分解性ポリエステルフィルムと低分子量ポリオレフィンとの接着性、耐水性をより改良するために、基材層の表面に下塗り剤としてイソシアネート系などの接着剤を生分解性ポリエステルフィルムに塗布しておいた方が好ましい。かかる下塗り剤の塗布量は、通常、固形分重量で５０～１５０ｍｇ／ｍ^{2}の範囲が好ましい。前記被覆用組成物の塗布量は、通常、固形分重量で０．１～５ｇ／ｍ^2、好ましくは０．５～２ｇ／ｍ^{2}の範囲にある。また上記下塗り剤の濡れを良くするため０．１～２５Ｗ／ｍ^2・分のコロナ放電処理を行うことが好ましい。

【００１９】低分子量ポリオレフィンの水性分散体を塗布した生分解性ポリエステル積層フィルムの乾燥温度は、通常、５０～１２０℃、好ましくは６０～９０℃の範囲がよい。乾燥温度が低いと、良好な被膜が形成されなかったり、乾燥時間を長くすることを必要とし、生産性が低下する虞がある。一方、乾燥温度が高すぎると、代表的な生分解性ポリエステルフィルムであるポリ乳酸等の耐熱性が低いフィルムは乾燥時に軟化してシワが発生する虞がある。また低分子量ポリオレフィンを融解し防湿膜とするには乾燥と同時又は乾燥後に１２０～１６０℃の熱処理を行う必要がある。かかる熱処理は生分解性ポリエステルフィルムに負荷を与えるため、オーブン等で端部を固定しながら行うのが望ましく、また紙またはデンプンシートにラミネートした後に容器成形の金型の熱で成形と同時に熱処理を行っても良い。

【００２０】

【発明の効果】本発明の生分解性ポリエステル積層フィルムは、高い機械的強度を有し、かつ優れた水蒸気ガスバリヤー性を有するので、高い水蒸気バリア性能が必要なため、従来ポリオレフィンフィルムしか使用できなかった乾物等の食品包装も可能となり、内容物（食品）も含めたコンポストごみの回収、処理が容易になり、生分解性プラスチック製品の用途を大きく広げることができる。その他発明の利用形態として、紙とのラミを用いた食品または廃棄用のコンポスト容器等が挙げられる。又、かかる生分解性ポリエステル積層フィルムを紙、パルプモールドまたはデンプン混練樹脂に積層して包装材料、トレー、容器等として利用することもできる。

【００２１】

【実施例】次に実施例を挙げて本発明を更に具体的に説明するが、本発明はその要旨を越えない限りこれらの実施例に制約されるものではない。

【００２２】実施例及び比較例で得られた積層体の物性値は以下の方法で測定した。

（１）光学特性

日本電色工業社製　ヘイズメーター３００Ａを用いてヘイズ、平行光線透過率（ＰＴ）及びグロスを測定した。

（２）透湿度

ＪＩＳ　Ｚ０２０８　防湿包装材料の透湿度試験（カップ法）による。

【００２３】実施例及び比較例で使用した原料は次の通りである。

（１）低分子量ポリエチレンの水性分散体（ＰＯ－１）

低分子量ポリエチレンの物性：密度：０．９２ｇ／ｃｍ^3、針入度法硬度（ＪＩＳ　Ｋ２２０７）：３、融点：１１０℃、粘度平均分子量：４０００水性分散体の物

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性：固形分濃度：４０重量%、ｐＨ：９、粘度：１００ＯＣＰ、粒径：４μｍ

（２）低分子量ポリエチレンの水性分散体（ＰＯ－２）

低分子量ポリエチレンの物性：密度：０．９５ｇ／ｃｍ^3、融点：１３２℃、粘度平均分子量：４０００

水性分散体の物性：固形分濃度：４０重量%、ｐＨ：９、粘度：１０００ＣＰ、粒径：７μｍ

（３）生分解性ポリエステル：以下の組成からなるスルホン酸金属塩基を有する芳香族ジカルボン酸を共重合体として含む生分解性ポリエステル樹脂を用いた。共重合体のモノマー組成：テレフタル酸４５モル%、エチレングリコール３７モル%、ジエチレングリコール９モル%、５－スルホーイソフタル酸ナトリウム１モル%、ヒドロキシ酢酸８モル%。共重合体の密度：１．３５（ｇ／ｃｍ^3）、融点：２００℃、メルトフローレート（２２０℃、２１６０ｇ荷重）：１５（ｇ／１０分）。

【００２４】生分解性ポリエステル二軸延伸フィルムの製造：ブルックナー社の逐次二軸延伸機を用いて製膜した。上記生分解性ポリエステル樹脂をオーブン中で予備乾燥し、押出機（６０ｍｍφ、シリンダー設定温度：２３０℃）中へ供給して溶融した。押出機先端に設けたＴダイからフィルム状に押出し、キャストロールで３０℃に冷却し、厚さ２５０μｍのシートを成形した。次に、逐次二軸延伸工程で、このシートを６０℃で３．０倍縦延伸し、次いで６０℃で３．２倍横延伸し、引き続き１８０℃で２０秒間熱処理（ヒートセット）を行った。その後、フィルム両面にコロナ放電処理を施し、厚さ２５μｍの生分解性ポリエステル二軸延伸フィルムを得た。

【００２５】実施例１

上記生分解性ポリエステル二軸延伸フィルムの片面にイソシアネート系接着剤（三井武田ケミカル株式会社製タケネートＡ－３：１ｇに同Ａ－３１０：１２ｇを酢酸エチル１８７ｇで希釈し固形分３．４%液にする）を、マイヤーバーを用いて固形分重量で１００ ｍｇ／ｍ^{2}塗布し、６０℃で１０秒間乾燥した。次いで、前記低分子量ポエチレン水性分散体（ＰＯ－１）をマイヤーバーを用いて固形分重量で ２．０ｇ／ｍ^{2}になるように塗布した後、ドライヤーを用いて７０℃で２０秒間乾燥し積層フィルムを得た。尚、それぞれの塗布液は界面活性剤として花王株式会社製ラウリルエーテルＰＯＥ（１３，３）商品名エマルゲン１２０を０．４ｗｔ%加えてある。得られた積層フィルムを厚み３ｍｍのガラス板に固定し四方を４ｃｍ間隔でクリップで固定し、熱による収縮を抑えて強制対流型高温槽（イズミ製作所製オーブン）に入れ、１６０℃×５分の条件で熱固定処理を行った。得られた積層フィルムの物性値を表１に示す。

【００２６】実施例２

実施例１で用いたは低分子量ポリエチレン水性分散体（ＰＯ－１）に代えて、低分子量ポリエチレン水性分散体（ＰＯ－２）を用いる以外は実施例１と同様に行い積層フィルムを得た。得られた積層フィルムの物性値を表１に示す。

【００２７】実施例３

実施例２で用いた低分子量ポリエチレン水性分散体（ＰＯ－２）に代えて、低分子量ポリエチレン水性分散体（ＰＯ－２）に更に１，４－ブタンジオール・コハク酸エステルの水性分散体（昭和高分子社製　商品名ビオノーレエマルジョンＯＬＸ－７５２７、融点：９３～９５℃、ガラス転移点：－４５℃、結晶化度：２０～３５%、比重：１．３、水生分散体の物性：固形分濃度：５０～５６重量%、ｐＨ：４～６、粘度：１００ＣＰ、粒径：１～２μｍ）を固形分中に１２重量%配合し、更に１００００ｒｐｍで１分間攪拌して得た水性分散体を用いる以外は実施例２と同様に行い積層フィルムを得た。得られた積層フィルムの物性値を表１に示す。

【００２８】比較例１

前記フィルム両面にコロナ放電処理した厚さ２５μｍの生分解性ポリエステル二軸延伸フィルムの物性値を比較例１として表１に示す。

【００２９】

【表１】

	実施例１	実施例２	実施例３	比較例１
低分子量ポリエチレン層	PO-1	PO-2	PO-2	なし
物性値				
透湿度 g/m^2・day	19	19	31	70
ヘイズ %	61	88	92	4
光線透過率 %	36	11	7	69

【００３０】表１に示した結果から明らかなように、低分子量ポリエチレン層が積層されてなる生分解性ポリエステル二軸延伸積層フィルム（実施例１～実施例３）が、低分子量ポリエチレン層を有しない生分解性ポリエステル二軸延伸フィルムに比べ、透湿度が改良されている。

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(54) LAMINATED FILM OF BIODEGRADABLE POLYESTER

(57)Abstract:
PROBLEM TO BE SOLVED: To provide a laminated film of a biodegradable polyester which has improved moistureproofness without marring biodegradability.
SOLUTION: The laminated film of the biodegradable polyester has a low molecular weight polyolefin layer preferably falling within the viscosity-average molecular weight range of 1,000 to 7,000, laminated on at least one side of a base material layer of a biodegradable polyester film comprising an aliphatic polyester such as polylactic acid or the like and also improved moistureproofness without marring biodegradability.

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(54)【発明の名称】　生分解性樹脂組成物

(57)【要約】
【課題】　優れた水崩壊性、及び生分解性を有する生分解性樹脂組成物を提供する。
【解決手段】　生分解性樹脂とポリエチレングリコールの合計量２５～８５重量%、及び無機充填剤７５～１５重量%を含む生分解性樹脂組成物であって、前記生分解性樹脂１００重量部に対して、数平均分子量が５０００～３００００であるポリエチレングリコール１０～１００重量部を含むことを特徴とする生分解性樹脂組成物。

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【特許請求の範囲】

【請求項１】　生分解性樹脂とポリエチレングリコールの合計量２５～８５重量%、及び無機充填剤７５～１５重量%を含む生分解性樹脂組成物であって、前記生分解性樹脂１００重量部に対して、数平均分子量が５０００～３００００であるポリエチレングリコール１０～１１０重量部を含むことを特徴とする生分解性樹脂組成物。

【請求項２】　生分解性樹脂が、乳酸系ポリマー、及びポリアルキレンサクシネートから選ばれた少なくとも一種の樹脂であることを特徴とする請求項１記載の生分解性樹脂組成物。

【請求項３】　乳酸系ポリマーがポリ乳酸であることを特徴とする請求項２記載の生分解性樹脂組成物。

【請求項４】　ポリアルキレンサクシネートが、ポリブチレンサクシネート、ポリブチレンサクシネート・アジペート、ポリブチレンサクシネート・テレフタレート、及びポリエチレンサクシネートから選ばれた少なくとも一種の樹脂であることを特徴とする請求項２記載の生分解性樹脂組成物。

【請求項５】　無機充填剤が、炭酸カルシウム、硫酸バリウム、タルク、クレー、シリカ、及び雲母から選ばれた少なくとも一種の化合物であることを特徴とする請求項１記載の生分解性樹脂組成物。

【請求項６】　生分解性樹脂組成物が、請求項１記載の生分解性樹脂組成物を成形加工したシート、フィルム、及び容器から選ばれた少なくとも一種の成形物であることを特徴とする生分解性樹脂組成物。

【請求項７】　成形物が、生理用タンポンのアプリケータ、生理用ナプキン包装用フィルム、使い捨ておむつ用バックシート、及び医療用衣類から選ばれた少なくとも一種の衛生用品であることを特徴とする請求項６記載の樹脂組成物。

【請求項８】　２３℃の水に２日間浸漬したときの重量減少率が少なくとも１０%であり、且つ２３℃の水に１０分間浸漬したときの重量減少率が１０%未満であることを特徴とする請求項１～７のいずれか１項に記載の生分解性樹脂組成物。

【発明の詳細な説明】

【０００１】

【発明の属する技術分野】本発明は、生分解性樹脂組成物に関する。詳しくは、生理用タンポン、ナプキン、紙おむつ、及び医療用衣類等の衛生用品の成形用材料に適し、使用時は体液等により崩壊することがなく通常と全く同様に使用することができ、使用後トイレ、台所流し等に放流して水と接触した後には、崩壊及び分解する特性を有する生分解性樹脂組成物に関する。

【０００２】

【従来の技術】従来、生理用タンポン、ナプキン、使い捨ておむつ等は、トイレ等から放流処理することを想定して設計されていない。本来であれば、これらの物品、特に生理用品はトイレ内で装着、脱着がされることが多く、使用後はトイレットペーパーなどと同様に放流処理したほうが、簡単で汚れず形跡も残らないため好ましい。

【０００３】しかしながら、これらの物品の大部分は、一般的にポリエチレン、ポリプロピレン等の非分解性樹脂からなっており、放流することが出来ないため、使用者は別途設けられた箱などへ入れるか、持ち帰って別途処理する煩わしさがある。また、その後、ゴミとしての処理する際に、飛散したり、埋め立て処理される場合に分解しない等の問題もある。さらに、万が一トイレにて放流された場合は、下水処理系に蓄積されたり閉塞を引き起こす。また、分解されないためしばしば河川、海洋へ放出され環境への悪影響が大きくなる。このように、使用者にとっては処理の煩わしさ、処理時に手が汚れる等の不衛生さ、処理の形跡が残る等の心理的圧迫からの開放が望まれている。また、処理面からは、衛生面、環境への負加を改善する必要性が唱えられている。

【０００４】これらの問題を解決する方法として、特開平１０－１３１０８３号公報等には、水解紙が開示されている。しかし、これらは、紙を原料として用いており、衛生用品等のように風合いが重視される用途には不向きであった。紙ではなくプラスチックを用いた例として、特開昭６３－３０２８４５号公報、特開平５－２９５２１１号公報、特開平６－１３４９１０号公報、特公平７－５７２３０号公報などに、水溶性樹脂を用いた衛生用品の例示がある。これらの開示によれば、ポリビニルアルコールやポリ－３－ヒドロキシ酪酸等の水溶性樹脂を用いる事により、上記問題の解決を図っている。

【０００５】また、特開平６－２９９０７７号公報には、生分解性プラスチックス２０～８０重量%と水溶性熱可塑性樹脂８０～２０重量%を混合して得られる生分解性樹脂組成物の開示されている。しかしながら、これらの水溶性樹脂や生分解性樹脂は一般的に高価であり、使用量、添加量を増やすと価格が高くなり、衛生用品のような使い捨て用途には好ましくない。逆に、安価にするため添加量を減らすと、水崩壊性、生分解性が不足し、放流処理が出来ず好ましくない。さらには、本発明者らの知見では、これらの水溶性樹脂を含む樹脂組成物は経時的に水分を吸収しやすく、使用前に湿潤して表面がべとべとになったり、カビが発生したりするため、実使用上問題がある場合がある。

【０００６】

【発明が解決しようとする課題】本発明者らは、上記問題点を解決する手段として、特願２００１－１１４７７２号に係わる特許出願において、生分解性樹脂２５～８０重量%、無機充填剤７５～２０重量%、及び、前記生分解性樹脂１００重量部に対して水溶性樹脂３～１００重量部を含むことを特徴とする生分解性樹脂組成物を提案した。該樹脂組成物は、良好な水崩壊性、生分解性を

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有し、使用時には経時的な表面状態の悪化がない成形物を与える。しかしながら、該組成物を実際の使用形態であるタンポン用アプリケータなどに成形して評価した場合、水への溶解速度が比較的速いため、使用時に体内に成分が若干残存する可能性があり、また、成形物の保管中、経時的に硬化が進行することがあり、用途によっては必ずしも満足できない点があることが判った。

【０００７】本発明の目的は、上記の問題点に鑑み、優れた水崩壊性、及び生分解性を有しながらも、実使用時には成分が溶け出すことがほとんどなく経時的にも安定で、使用後、トイレ、台所流し等へ放流処理するだけで簡単に崩壊する生分解性樹脂組成物を提供することにある。

【０００８】

【課題を解決するための手段】本発明者らは、上記課題を達成するため鋭意検討した結果、特定量の生分解性樹脂、特定量の無機充填剤、及び特定量のポリエチレングリコールを含む生分解性樹脂組成物が、良好な水崩壊性、生分解性を有し、使用時には成分の溶けだしがほとんどなく、経時的にも安定な成形物を与えることを見出し、本発明を完成するに至った。

【０００９】すなわち、本発明は、生分解性樹脂とポリエチレングリコールの合計量２５～８５重量%、及び無機充填剤７５～１５重量%を含む生分解性樹脂組成物であって、前記生分解性樹脂１００重量部に対して、数平均分子量が５０００～３００００であるポリエチレングリコール１０～１１０重量部を含むことを特徴とする生分解性樹脂組成物である。

【００１０】本発明に係わる生分解性樹脂組成物の好ましい態様として、上記生分解性樹脂が、乳酸系ポリマー、及びポリアルキレンサクシネートから選ばれた少なくとも一種の樹脂である前記樹脂組成物が挙げられる。それらの内、乳酸系ポリマーとしてはポリ乳酸、ポリアルキレンサクシネートとしては、ポリブチレンサクシネート、ポリブチレンサクシネート・アジペート、ポリブチレンサクシネート・テレフタレート、及びポリエチレンサクシネートから選ばれた少なくとも一種の樹脂が挙げられる。また、上記無機充填剤として、炭酸カルシウム、硫酸バリウム、タルク、クレー、シリカ、及び雲母から選ばれた少なくとも一種の化合物が挙げられる。

【００１１】本発明に係わる前記各種の生分解性樹脂組成物は、これらを成形加工することにより、シート、フィルム、容器、又はこれらの組み合わせ成形物となすことができる。具体的成形物として、生理用タンポンのアプリケータ、生理用ナプキン包装用フィルム、使い捨ておむつ用バックシート、及び医療用衣類から選ばれた少なくとも一種の成形物が挙げられる。これらは、トイレ又は台所流しへ放流処理可能な衛生用品である。

【００１２】本発明の生分解性樹脂組成物の主な特性としては、２３℃の水に２日間浸漬したときの重量減少率が少なくとも１０%であり、且つ２３℃の水に１０分間浸漬したときの重量減少率が１０%未満であることが挙げられる。

【００１３】本発明の生分解性樹脂組成物は、特定量の生分解性樹脂、ポリエチレングリコール、及び無機充填剤を含むために、優れた水崩壊性、生分解性を有し、経時的にも表面状態の悪化がなく、しかも廉価である。また、使用時に成分が溶け出すことがなく、経時的にも安定である。そのため、上記衛生用品などの成形物として用いた場合、使用後の処理が非常に簡便で衛生的となり、処理後も環境への負加を減らすことが可能である。

【００１４】

【発明の実施の形態】以下、本発明について詳しく説明する。本発明の生分解性樹脂組成物は、特定量の生分解性樹脂、無機充填剤、及びポリエチレングリコールを混合することにより製造される。上記材料を混合した後、連続溶融混練を行うことが好ましい。

【００１５】本発明に用いる生分解性樹脂としては、乳酸系ポリマー、ポリブチレンサクシネート、ポリブチレンサクシネート・アジペート、ポリブチレンサクシネート・テレフタレート、ポリエチレンサクシネート、及びポリブチレンサクシーネート・カーボネート等のポリアルキレンサクシネート、ポリグリコール酸、ポリカプロラクトン、ポリヒドロキシ酪酸、ポリヒドロキシ吉草酸、ヒドロキシ酪酸・ヒドロキシ吉草酸共重合体等が挙げられる。これらの内、乳酸系ポリマー、ポリブチレンサクシネート、ポリブチレンサクシネート・アジペート、ポリブチレンサクシネート・テレフタレート、及びポリエチレンサクシネートが好ましい。

【００１６】上記乳酸系ポリマーとしては、ポリ乳酸、または乳酸と他のヒドロキシカルボン酸とのコポリマーが挙げられる。コモノマーとして用いられる他のヒドロキシカルボン酸として、グリコール酸、ヒドロキシ酪酸、ヒドロキシ吉草酸、ヒドロキシペンタン酸、ヒドロキシカプロン酸、ヒドロキシヘプタン酸等が例示される。

【００１７】好ましいポリ乳酸の分子構造は、Ｌ－乳酸またはＤ－乳酸いずれかの単位８５～１００モル%、さらに好ましくは該単位８５～９８モル%、及び、それぞれの対掌体の乳酸単位０～１５モル%、さらに好ましくは、該対掌体単位２～１５モル%を含むものである。また、乳酸と他のヒドロキシカルボン酸とのコポリマーは、Ｌ－乳酸またはＤ－乳酸いずれかの単位８５～１００モル%、好ましくは該単位８５～９８モル%、及び、他のヒドロキシカルボン酸単位０～１５モル%、好ましくは該単位２～１５モル%を含むものである。好ましい他のヒドロキシカルボン酸として、グリコール酸、ヒドロキシカプロン酸が挙げられる。

【００１８】これらの乳酸系ポリマーは、Ｌ－乳酸、Ｄ－乳酸および他のヒドロキシカルボン酸の中から必要と

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する構造のものを選んで原料とし、脱水重縮合することにより得ることができる。好ましくは、乳酸の環状二量体であるラクチド、グリコール酸の環状二量体であるグリコリド、およびカプロラクトン等から必要とする構造のものを選んで開環重合することにより得ることができる。

【００１９】ラクチドには、Ｌ－乳酸の環状二量体であるＬ－ラクチド、Ｄ－乳酸の環状二量体であるＤ－ラクチド、Ｄ－乳酸とＬ－乳酸とが環状二量化したメソ－ラクチド、およびＤ－ラクチドとＬ－ラクチドとのラセミ混合物であるＤＬ－ラクチドがある。本発明ではいずれのラクチドも用いることができる。ただし、主原料は、Ｄ－ラクチドまたはＬ－ラクチドが好ましい。

【００２０】本発明に用いられる好ましい乳酸系ポリマーは、上記組成のポリ乳酸、または乳酸と他のヒドロキシカルボン酸とのコポリマーであるが、それらは、次の（１）～（６）の方法により得ることができる。

（１）Ｌ－ラクチドをおよそ８５モル％以上とＤ－ラクチドおよび、またはグリコリドをおよそ１５モル％以下とを共重合させる、（２）Ｄ－ラクチドをおよそ８５モル％以上とＬ－ラクチドおよび、またはグリコリドをおよそ１５モル％以下とを共重合させる、（３）Ｌ－ラクチドをおよそ７０モル％以上とＤＬ－ラクチドおよび、またはグリコリドをおよそ３０モル％以下とを共重合させる、（４）Ｌ－ラクチドをおよそ７０モル％以上とメソ－ラクチドおよび、またはグリコリドをおよそ３０モル％以下とを共重合させる、（５）Ｄ－ラクチドをおよそ７０モル％以上とＤＬ－ラクチドおよび、またはグリコリドをおよそ３０モル％以下とを共重合させる、

（６）Ｄ－ラクチドをおよそ７０モル％以上とメソ－ラクチドおよび、またはグリコリドをおよそ３０モル％以下とを共重合させる。

【００２１】これらの乳酸系ポリマーは高分子量であることが好ましく、濃度０．５ｇ／ｄｌのクロロホルム溶液（２５℃）の固有溶液粘度が１～１０であることが好ましく、さらに好ましくは３～７である。固有溶液粘度が１未満であると、溶融粘度が低すぎて押出機のダイのスリットから流下し、成形し難いばかりでなく、脆すぎて取扱が困難であり、また、１０を超えると溶融粘度が高過ぎて押出成形性等が悪くなるので好ましくない。

【００２２】ラクチド、または、ラクチドとグリコリドを重合させて、短時間で高分子量のポリマーを得るためには触媒を用いるのが好ましい。このような重合触媒としては、この重合反応に触媒効果を示す各種のものが使用できる。例えば、公知なものとして、オクタン酸第一錫、四塩化錫、塩化亜鉛、四塩化チタン、塩化鉄、三フッ化ホウ素エーテル錯体、塩化アルミニウム、三フッ化アンチモン、酸化鉛等の主として多価金属を含む化合物が挙げられ、中でも錫化合物または亜鉛化合物が好ましく使用される。錫化合物の中ではオクタン酸第一錫が特に好ましい。使用量はラクチド、または、ラクチドとグリコリドの総量に対して、およそ０．００１～０．１重量％程度が好ましい。

【００２３】また、重合の際には、公知の連鎖増大剤を用いることができる。連鎖増大剤としては、ラウリルアルコール等の高級アルコール類、乳酸やグリコール酸等のヒドロキシ酸類が好ましく用いられる。連鎖増大剤の共存により、重合速度が大きくなるので短時間でポリマーを得ることができる。また、連鎖増大剤の量を加減することによりポリマーの分子量を調節することもできる。しかし、連鎖増大剤の量を多くしすぎると生成ポリマーの分子量は小さくなる傾向があるので、連鎖増大剤を使用する場合には、その量はラクチド、または、ラクチドとグリコリドの総量に対して０．１重量％以下であることが好ましい。

【００２４】重合または共重合するに当たって、溶媒を用いても用いなくてもよいが、高分子量のポリマーを得るには、ラクチドまたはグリコリドを溶融した状態で塊状重合することが好ましい。重合温度は、溶融重合の場合には原則的にはモノマーであるラクチド、またはラクチドとグリコリドの融点（９０℃付近）以上の温度であればよい。また、例えば、クロロホルム等の溶媒を用いる溶液重合の場合には、ラクチド、または、ラクチドとグリコリドの融点以下の温度で重合することが可能である。いずれの場合も、２５０℃を超えると生成ポリマーの分解が起こるので好ましくない。ポリ乳酸の代表的な市販品として、三井化学（株）製、商品名：レイシアが挙げられる。

【００２５】ポリアルキレンサクシネートとしては、ポリブチレンサクシネート、ポリブチレンサクシネート・アジペート、ポリブチレンサクシネート・テレフタレート、ポリエチレンサクシネート、及びポリブチレンサクシーネート・カーボネートが好ましい。ポリブチレンサクシネートの市販品として、昭和高分子（株）製、商品名：ビオノーレ１００１、同ビオノーレ１０２０、同ビオノーレ１９０３等が挙げられる。また、ポリブチレンサクシネート・アジペートの市販品として、昭和高分子（株）製、商品名：ビオノーレ３００１、同ビオノーレ３０２０等が挙げられる。

【００２６】ポリブチレンサクシネート・テレフタレートの市販品として、ＢＡＳＦ社製、商品名：エコフレックス、ＤｕＰｏｎｔ社製、商品名：バイオマックス等が挙げられる。ポリエチレンサクシネートの市販品として、（株）日本触媒製、商品名：ルナーレＳＥが挙げられる。ポリブチレンサクシネート・カーボネートの市販品として、三菱ガス化学（株）製、商品名：ユーペックが挙げられる。

【００２７】本発明の生分解性樹脂組成物は、上記生分解性樹脂１００重量部に対し可塑剤を１～５０重量部含んでもよい。可塑剤として、例えば、ジーｎ－オクチル

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フタレート、ジー２－エチルヘキシルフタレート、ジベンジルフタレート等のフタル酸誘導体、ジイソオクチルフタレート等のイソフタル酸誘導体、ジーｎ－ブチルアジペート、ジオクチルアジペート等のアジピン酸誘導体、ジーｎ－ブチルマレート等のマレイン酸誘導体、トリーｎ－ブチルシトレート等のクエン酸誘導体、モノブチルイタコネート等のイタコン酸誘導体、ブチルオレート等のオレイン酸誘導体、グリセリンモノリシノレート等のリシノール酸誘導体、トリクレジルフォスフェート、トリキシレニルフォスフェート等のリン酸エステル系可塑剤、アセチルクエン酸トリエチル、アセチルクエン酸トリブチル、乳酸、直鎖状乳酸オリゴマー、環状乳酸オリゴマーまたはラクチド等が例示できる。特に分子内に２個以上のカルボン酸エステル基を有する、クエン酸エステル、グリセリンエステル、フタル酸エステル、アジピン酸エステル、セバシン酸エステル、アゼライン酸エステルおよびトリエチレングリコールエステルから選ばれた少なくとも１種のエステル化合物であることが好ましい。これらの可塑剤は単独でも良いし、また２種以上を混合して用いてもよい。

【００２８】かかる可塑剤を乳酸系ポリマーに添加することにより、乳酸系ポリマーは効果的に可塑化され、得られる樹脂組成物は柔軟性を帯びる。樹脂組成物中の可塑剤の量が１重量％以上になると柔軟性がはっきり現れるようになり、５０重量％を超えると、該組成物を溶融押出する時の成形性が悪くなり、また、得られた成形物の強度が弱くなったり、経時的に可塑剤が浮き出してくるため好ましくない。

【００２９】本発明の生分解性樹脂組成物には、上記生分解樹脂と、特定量の無機充填剤、及び生分解性樹脂に対し特定量のポリエチレングリコールが添加される。ポリエチレングリコールとしては、ポリエチレングリコール、またはその変性物が挙げられる。ポリエチレングリコールの数平均分子量は、本発明の樹脂組成物を成形した際の、水への溶解速度、及び経時的な安定性に影響を及ぼす。数平均分子量が低すぎると、水への溶解速度が速すぎ、例えばタンポンアプリケータとして用いた場合、使用時に体内に成分が残存したり、極端な場合、体内で崩壊が進行する可能性が大きくなる。また、成形後に保管する際、経時的に樹脂が硬化するなどの問題が生じる。

【００３０】かかる点を考慮すると、ポリエチレングリコールの数平均分子量は５０００以上であることが好ましい。更に好ましくは７０００以上、特に好ましくは９０００以上である。数平均分子量の上限については、高ければ高いほど好ましい。原料の入手の容易なことを考慮すると、通常、数平均分子量の上限は３００００程度である。好ましくは２００００程度である。上記ポリエチレングリコールは数平均分子量の異なるものを混合しても良い。混合前の個別の数平均分子量が５０００以下であっても、混合後の全体としての数平均分子量（以下、Ｍｎという）が５０００以上であれば問題はない。

【００３１】ポリエチレングリコールの市販品として、三洋化成工業（株）製、商品名：ＰＥＧ－１０００（Ｍｎ：１０００）、同－４０００Ｓ（Ｍｎ：３３００）、同－６０００Ｓ（Ｍｎ：８３００）、同－１００００（Ｍｎ：１１０００）、同－１３０００（Ｍｎ：１３０００）、同－２００００（Ｍｎ：２００００）等が挙げられる。ポリエチレングリコールのＭｎの算出方法としては、まず無水酢酸・ピリジン法により水酸基価を求め、その水酸基価からＭｎを算出する。詳細については後述の実施例に記載する。

【００３２】本発明の生分解性樹脂組成物は、上記生分解性樹脂に対し、特定量の上記ポリエチレングリコールを含む。ポリエチレングリコールの含有量は、樹脂組成物の水崩壊性、生分解性に影響を及ぼす。水溶性樹脂の含有量が少な過ぎると、水崩壊性が不足し、その結果、生分解性も抑制されることとなり好ましくない。逆に、水溶性樹脂の含有量が多過ぎると、水崩壊性は良好であるが、樹脂組成物の水親和性が過剰となり、水と隔離された状態であっても、空気中の湿気により経時的に成形物の表面がべとべとしたり、カビが発生したりすることがある。また、樹脂組成物が高価となるため、廉価性が必要とされる衛生用品のような使い捨て用途には好ましくない。さらに、結果として相対的に生分解性樹脂の量が低下するため、強度が低下する問題がある。かかる点を考慮すると、生分解性樹脂１００重量部に対して、ポリエチレングリコール１０～１１０重量部を含むことが好ましい。より好ましい添加量としては１５～１０５重量部である。

【００３３】本発明に用いられる無機充填剤としては、炭酸カルシウム、炭酸マグネシウム、炭酸バリウム、硫酸マグネシウム、硫酸バリウム、硫酸カルシウム、酸化亜鉛、酸化マグネシウム、酸化カルシウム、酸化チタン、酸化マグネシウム、アルミナ、水酸化アルミニウム、ヒドロキシアパタイト、シリカ、マイカ、タルク、カオリン、クレー、ガラス粉、アスベスト粉、ゼオライト、珪酸白土、雲母等が挙げられる。これらの内、炭酸カルシウム、酸化マグネシウム、硫酸バリウム、シリカ、珪酸白土等が好ましく用いられる。より好ましくは、炭酸カルシウム、硫酸バリウムである。これらの無機充填剤は単独で用いても良いし、２種以上混合して用いても良い。

【００３４】上記無機充填剤と生分解性樹脂との配合比は、得られる樹脂組成物の分解性、表面状態、廉価性に影響を及ぼす。無機充填剤の含有量が多過ぎると、樹脂成分が極端に低下し強度が不足するため好ましくない。また、樹脂への混練も困難となり生産性が低下する。逆に少な過ぎると、強度や混練の難易性には問題がないが、水崩壊性が低下する上に、樹脂組成物の廉価性が損

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なわれるために好ましくない。また、無機充填剤は多少なりとも吸着性があり、樹脂組成物が空気中の湿気により経時的に表面がべとべとしたり、カビが発生したりするのを防ぐ効果があるが、その効果が低下するために好ましくない。かかる観点から、本発明の生分解性樹脂組成物は、生分解性樹脂とポリエチレングリコールの合計量２５～８５重量%、及び無機充填剤７５～１５重量%を含むことが好ましい。より好ましくは、生分解性樹脂とポリエチレングリコールの合計量３０～９０重量%、無機充填剤７０～１０重量%である。

【００３５】これらの無機充填剤の平均粒径としては、０．３～１０μmのものが好ましい。さらに好ましくは０．５～５μmである。平均粒径が１０μmを超えると樹脂組成物の表面状態が荒くなる上に、美観も損ねるため好ましくない。また、フィルムやシートなどへの成形の際、穴があくなどの不良が発生しやすく生産性、作業性等の点でも好ましくない。平均粒径が０．３μm未満の場合、無機充填剤の凝集が発生しやすく、均一な練り込みが出来ないため好ましくない。また、無機充填剤の嵩密度が低くなり、混練時の作業性が低下し生産性が低下するため好ましくない。

【００３６】無機充填剤は、樹脂との分散性を向上させるために表面処理を行っても良い。但し、表面処理により無機充填剤の吸湿性が損なわれ、樹脂組成物の経時的な表面状態の悪化を抑制できない場合はこの限りではない。表面処理剤としては、例えば、ステアリン酸、ラウリン酸等の高級脂肪酸またはそれらの金属塩等を挙げることができる。本発明の生分解性樹脂組成物には、その他に本発明の目的を妨げない範囲で分散剤、顔料、安定剤、酸化防止剤、着色剤、紫外線吸収剤等の他の添加剤を添加してもよい。

【００３７】上記の通り、本発明の生分解性樹脂組成物は、特定量の生分解性樹脂、ポリエチレングリコール、及び無機充填剤を含み、優れた水崩壊性、生分解性樹脂を有し、経時的にも表面状態の悪化がなく、しかも廉価である。優れた水崩壊性及び生分解性を有する理由は定かではないが、一応、次ぎのような機構を推定する。

【００３８】本発明の生分解性樹脂組成物が水中あるいは土壌中に廃棄された場合、先ず、樹脂組成物中のポリエチレングリコールが水に溶解する。その結果、樹脂組成物中には空隙が多くでき、ポリエチレングリコールの溶解が進むにつれて破断され、場合によっては粒状にまで崩壊する。本発明の生分解性樹脂組成物には無機充填剤が多く含まれているため、この崩壊の際に同時に無機充填剤が脱落することにより、ますます崩壊が促進される。その結果、樹脂組成物中には空隙が多く形成され、樹脂の表面積が増えることとなり、残った生分解性樹脂は比較的速やかに分解するものと推定される。

【００３９】次いで、本発明の生分解性樹脂組成物の製造方法を例示する。例えば、生分解性樹脂、無機充填剤、ポリエチレングリコール、必要に応じて可塑剤、分散剤、安定剤などその他の添加物をヘンシェルミキサー、スーパーミキサー、タンブラー型ミキサー等を用いて混合した後、一軸または二軸スクリュー型押出機を用いて連続混練する。ここで、生分解性樹脂中への無機充填剤、ポリエチレングリコールの分散性をより向上させるためには、二軸押出機の方が好ましい。

【００４０】押出温度は、好ましくは１００～２７０℃の範囲、更に好ましくは１３０～２５０℃の範囲である。１００℃未満では、押出安定性が得難く、また過負荷に陥りやすく、２７０℃を超えると、生分解性樹脂やポリエチレングリコールの分解が激しくなるので好ましくない。

【００４１】本発明の生分解性樹脂組成物から成形し得る成形体としては、例えば、生理用タンポン用アプリケータ、生理用ナプキン包装用フィルム、生理用ナプキン用バックシート、使い捨てオムツ用バックシート、使い捨てオムツ包装用フィルム等の衛生用品の容器、フィルム、シートが挙げられる。例えば、生理用タンポンアプリケータは、直径が２～３０mm、長さが１０～２００mm、厚みが０．０５～５mmの筒状容器であって、射出成形で成形したり、シート状物を得た後、筒状に丸める等により成形できる。

【００４２】また、フィルム、シートを得る場合には、Ｔダイなどが装着された押出機により溶融押出成形することにより、厚み０．０１～５mmに成形可能である。これらの成形物は、その分解速度を高めるために薄く成形することが好ましいが、強度等を満足させるように自由に調整可能である。好ましい厚みは、タンポン用アプリケータとしては０．１～５mm厚、より好ましくは０．２～２mm厚である。また、フィルムとしては、０．０１～０．１mm厚、より好ましくは０．０２～０．０７mm厚である。

【００４３】上記製造方法により得られる本発明の生分解性樹脂組成物、及びその成形物は、２３℃の水に２日間浸漬したときの重量減少率が少なくとも１０%であり、且つ２３℃の水に１０分間浸漬したときの重量減少率が１０%未満である。該２日間浸漬したときの重量減少率の上限は特に制限されるものではないが、本発明の範囲では主に生分解性樹脂や無機充填剤の配合比に影響され、約９０%程度である。該２日間浸漬したときの重量減少率が１０%未満であるような従来の樹脂組成物は、水崩壊性が不十分であり、上記条件を経過した後の崩壊、分解が極めて長期間に及ぶため好ましくない。より好ましくは、２日間浸漬したときの重量減少率が少なくとも１２%である。

【００４４】さらに、該２３℃の水に１０分間浸漬したときの重量減少率は１０%未満である。１０分間浸漬したときの重量減少率が１０%以上の場合、水への溶解速度は速すぎるため、例えばタンポンアプリケータなどに

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用いた場合、体内に成分が残存したり、極端な場合体内で崩壊が進む可能性が高くなり好ましくない。より好ましくは、１０分間浸漬したときの重量減少率は８％未満である。

【００４５】該２日間浸漬したときの重量減少率、及び１０分間浸漬したときの重量減少率は、ポリエチレングリコールの数平均分子量、及びその含有率やその他の添加剤も含んだ含有率と大きな相関がある。これらの３種の材料を本発明で規定する範囲で含むことにより、優れた水崩壊性、及び生分解性樹脂が発現するのである。その上、経時的にも表面状態の悪化及び樹脂の硬化などの現象がなく、実使用時には成分が溶け出すことがほとんどないため、使い捨ての衛生材料として極めて有用である。

【００４６】

【実施例】以下、実施例を示して本発明についてさらに詳細に説明する。なお、この実施例で用いた評価方法は、以下の通りである。

【００４７】（１）表面状態

実施例で得られた樹脂シートから、縦横それぞれ１０ｃｍ、厚み０．５ｍｍの試料を調製する。試料を４０℃、相対湿度６０％の恒温恒湿槽に２４時間放置した後の表面状態を観察する。評価基準は以下の通りである。○：表面状態の変化はほとんど認められない。ヌメリ感なし。△：表面状態の変化が若干認められる。ヌメリ感がある。×：表面がべとついている。

【００４８】（２）２日間重量減少率（％）

前項（１）と同様の試料を室温において、２３℃の純水５リットル中に２日間浸漬した後に取り出す。取り出した後、水道流水にて表面のヌメリ等がなくなるまで洗う。洗浄後、室温で２時間傾斜放置し水切りを行う。その後、シリカゲルを入れたデシケータ中で１２時間乾燥させる。下記数式（１）により重量減少率を算出する。１０枚の試料について測定し、その平均値を算出する。

$W_R = [(W_1 - W_2) / W_1] \times 100$ ・・（１）

ここで、W_R：重量減少率（％）、W_1：浸漬前の重量、W_2：浸漬後の重量。

【００４９】（３）１０分間重量減少率（％）

前項（１）と同様の試料を室温において、２３℃の純水５リットル中に１０分間浸漬した後に取り出す。取り出した後、室温で２時間傾斜放置し水切りを行う。その後、シリカゲルを入れたデシケータ中で１２時間乾燥させる。重量減少率を前項（２）と同様の方法で算出する。

【００５０】（４）無機充填剤の平均粒径（μｍ）

レーザー回折散乱法粒度分布測定器（日機装（株）製、形式：マイクロトラックＸ－１００）を用いて計測し、平均粒径（μｍ）を測定する。

【００５１】（５）経時強度

実施例で得られた試料（厚み０．５ｍｍのシート）を５０℃のオーブンに４日間放置したのち、１８０°に折り曲げ、下記の基準に従って評価を行う。

○：折り曲げ箇所が破壊せず、亀裂が生じない。△：折り曲げ箇所に亀裂が生じる。×：２つ以上の断片に破壊する。

【００５２】（６）ポリエチレングリコールの数平均分子量

水酸基価を測定し、得られた水酸基価から数平均分子量を算出する。

＜水酸基価（ＫＯＨｍｇ／ｇ）の測定＞［表１］に基づき、試料を三角フラスコに精秤し、無水酢酸・ピリジン混合試薬（無水酢酸１容にピリジン４容の割合で混合したもの）５ｍｌをホールピペットにて加える。９５～１００℃の油浴に上記三角フラスコを浸け、冷却器を付け１時間反応する。イオン交換水１ｍｌを加え、よく撹拌し、さらに１０分間油浴上で放置する。油浴から取り出し、さらに１０分間放置する。０．５％フェノールフタレインを指示薬として、０．５ｍｏｌ／Ｌ水酸化カリウム標準溶液で滴定する。以上の操作を２回行うと同時に、併行して空試験も行う。以下の式に従い水酸基価を求め、２回の平均をとる。

$OH = [\{(B - A) \times 28.05 \times f\} / S]$

ここで、ＯＨ：水酸基価（ＫＯＨｍｇ／ｇ）、Ｂ：空試験に要した０．５ｍｏｌ／Ｌ水酸化カリウム標準溶液のｍｌ数、Ａ：本試験に要した０．５ｍｏｌ／Ｌ水酸化カリウム標準溶液のｍｌ数、ｆ：０．５ｍｏｌ／Ｌ水酸化カリウム標準溶液の力価、Ｓ：試料採取量（ｇ）。

【００５３】尚、試料が遊離酸を含むときは、ＪＩＳＫ－１５５７－１９７０に規定される方法により酸価（ＫＯＨｍｇ／ｇ）を測定して、次の式により修正する。

修正ＯＨ＝ＯＨ＋酸価

ここで、修正ＯＨ：修正水酸基価（ＫＯＨｍｇ／ｇ）、ＯＨ：滴定で得られた水酸基価（ＫＯＨｍｇ／ｇ）。

【００５４】

【表１】

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水酸基価（KOHmg／g）	試料採取量（g）
５０未満	３．０～５．０
５０以上１００未満	２．０～３．０
１００以上１５０未満	１．５～２．０
１５０以上２００未満	１．０～１．５
２００以上２５０未満	０．８～１．０
２５０以上３００未満	０．６～０．８
３００以上４００未満	０．５～０．６

【００５５】＜数平均分子量の算出＞下記数式により算出する。

Ｍｎ＝（５６１００×２）／０Ｈ

ここで、Ｍｎ：数平均分子量、ＯＨ：滴定で得られた水酸基価又は上記修正水酸基価（ＫＯＨｍｇ／ｇ）。

調製例１

＜ジグリセリンテトラアセテートの調製＞ジグリセリン８４ｇ、及び無水酢酸４１５ｇを反応フラスコに入れ、強酸性イオン交換樹脂（ダウケミカル社製、商品名：ダウエックスＭＳＣ－１）２．５ｇを追加し、撹拌しながら８０～９０℃で１時間反応を行った。冷却後、イオン交換樹脂を濾別し、生成した酢酸および未反応の無水酢酸を減圧留去し、ジグリセリンテトラアセテート１６１．５ｇを得た。これは、一般式（１）におけるR_1～R_4が全て炭素数２のアシル基、ｎは１である、酸価が０．２、水酸基価が３．１である。

【００５６】実施例１～１６、比較例１～８

生分解性樹脂、可塑剤、ポリエチレングリコール、無機充填剤を〔表２〕～［表４］に示す銘柄、割合にて混合し、分散剤としてステアリン酸カルシウム（日東化成（株）製、商品名：Ｃａ－Ｓｔ）を上記樹脂組成物１００重量部に対して１重量部加え、ヘンシェルミキサーを用いて室温で混合して生分解性樹脂組成物を製造した。得られた樹脂組成物を１９０℃において、２軸スクリュー押出機にて混練した。さらに、混練物を１８０℃、１０ＭＰａにおいてプレス後、３０℃にてクエンチして、厚さ０．５ｍｍのシート状に加工した。得られたシートについて上記方法に従って物性を評価し、その結果を〔表２〕～［表４］に示す。

【００５７】

【表２】

		実施例1	実施例2	実施例3	実施例4	実施例5	実施例6	実施例7	実施例8
樹脂	種類	PLA	PLA	PLA	PLA	PLA	PLA	PLA	PLA
	量（重量部）	50	50	50	50	44	42	42	40
PEG	種類	6000S	10000	13000	20000	10000	10000	20000	20000
	Mn	8300	11000	13000	20000	11000	11000	20000	20000
	量（重量部）	14	14	14	14	14	22	22	22
	対樹脂100(重量部)	28	28	28	28	32	52	52	55
可塑剤	種類	－	－	－	－	－	－	－	－
	量（重量部）	－	－	－	－	－	－	－	－
無機充填剤	種類	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$BaSO_4$
	量（重量部）	36	36	36	36	42	36	36	38
２日間重量減少率	（重量%）	13	13	12	11	13	21	18	18
１０分間重量減少率	（重量%）	6	4	3	0	3	6	1	1
経時強度	－	△	○	○	○	○	○	○	○
表面状態	－	○	○	○	○	○	○	○	○

【００５８】

【表３】

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		実施例9	実施例10	実施例11	実施例12	実施例13	実施例14	実施例15	実施例16
樹脂	種類	PLA	PLA	PLA	PLA	PLA	PBS	PLA/m-PBT	PLA
	量(重量部)	40	70	30	47	50	50	25/25	40
PEG	種類	20000	20000	20000	10000	10000	10000	10000	20000
	Mn	20000	20000	20000	11000	11000	11000	11000	20000
	量(重量部)	35	13	14	20	14	14	14	40
	対樹脂100(重量部)	88	19	47	43	28	28	28	100
可塑剤	種類	−	−	−	A	−	−	−	−
	量(重量部)	−	−	−	3	−	−	−	−
無機充填剤	種類	$CaCO_3$	$CaCO_3$	$BaSO_4$	$CaCO_3$	talc	$CaCO_3$	$CaCO_3$	$CaCO_3$
	量(重量部)	25	17	56	30	36	36	36	20
2日間重量減少率	(重量%)	28	11	12	19	13	13	13	35
10分間重量減少率	(重量%)	2	1	1	5	3	4	4	4
経時強度	−	○	○	○	○	○	○	○	○
表面状態	−	○	○	○	○	○	○	○	○

【0059】 【表4】

		比較例1	比較例2	比較例3	比較例4	比較例5	比較例6	比較例7	比較例8
樹脂	種類	PLA	PLA	PLA	PLA	PLA	PLA	PLA	PLA
	量(重量部)	50	50	42	42	60	15	50	25
PEG	種類	1000	4000S	1000	4000S	10000	10000	10000	10000
	Mn	1000	3300	1000	3300	11000	11000	11000	11000
	量(重量部)	14	14	22	22	35	5	1	39
	対樹脂100(重量部)	28	28	52	52	58	33	2	156
可塑剤	種類	−	−	−	−	−	−	−	−
	量(重量部)	−	−	−	−	−	−	−	−
無機充填剤	種類	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$	$CaCO_3$
	量(重量部)	36	36	36	36	5	80	49	36
2日間重量減少率	(重量%)	14	14	22	22	30	−	0	33
10分間重量減少率	(重量%)	11	8	18	12	8	−	0	10
経時強度		×	×	×	×	○	−	×	×
表面状態		×	△	×	△	×	−	○	×

【0060】<表2～表4の記載の説明>PLA：ポリ乳酸（三井化学（株）製、商品名：レイシア）、PBS：ポリブチレンサクシネート（昭和高分子（株）製、商品名：ビオノーレ1001）、m－PBT：ポリブチレンサクシネート・テレフタレート（BASF社製、商品名：エコフレックス）、A：可塑剤（調製例1で得られたジグリセリンテトラアセテート）、PEG：ポリエチレングリコール［三洋化成工業（株）製、商品名：PEG－1000（Mn；1000）、同－4000S（Mn；4000）、同－6000S（Mn；6000）、同－10000（Mn；10000）、同－13000（Mn；13000）、同－20000（Mn；20000）］、$CaCO_3$：重質炭酸カルシウム（同和カルフィン（株）製、商品名：SST－40、平均粒径2μm）、$BaSO_4$：沈降性硫酸バリウム（バライト工業（株）製、商品名：ST、平均粒径0．9μm）。

【0061】実施例17

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実施例１～１６で得られた、厚み０．５ｍｍの各シートを筒状に丸めて、直径１０ｍｍ、長さ５０ｍｍの筒状の生理用タンポンアプリケータを製造した。

【００６２】

【発明の効果】本発明により提供される生分解性樹脂組成物は、優れた水崩壊性及び生分解性を有し、経時的にも表面状態の悪化がなく、しかも廉価である。そのため、本発明の生分解性樹脂組成物からの成形物は、使用時は体液等により崩壊することがなく通常と全く同様に使用でき、使用した後トイレ、台所流し等に放流した場合、水崩壊及び生分解する効果を奏する。その結果、環境への負加を減らすことが可能である。従って、生理用タンポンのアプリケータ、生理用ナプキン包装用フィルム、使い捨て紙おむつのバックシート、及び医療用衣類等の衛生用品や、キッチンタオル、掃除用タオル、ワイピングクロス等の生活用品の分野において好適に使用される。

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(54) BIODEGRADABLE RESIN COMPOSITION

(57)Abstract:
PROBLEM TO BE SOLVED: To provide a biodegradable resin composition having an excellent disintegrable property in water and an excellent biodegradable property.
SOLUTION: This biodegradable resin composition comprising a biodegradable resin and polyethylene glycol in a total amount of 25 to 85 wt.% and an inorganic filler in an amount of 75 to 15 wt.% is characterized in that the polyethylene glycol having a number-average molecular weight of 5,000 to 30,000 is contained in an amount of 10 to 100 pts.wt. per 100 pts.wt. of the biodegradable resin.

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Die folgenden Angaben sind den vom Anmelder eingereichten Unterlagen entnommen

(54) Ein- oder mehrschichtige, biologisch abbaubare, thermoplastische Folie mit verbesserten Barriereeigenschaften gegenüber Geruchsstoffen und deren Verwendung in Verpackungsmaterialien und Hygiene- und Kosmetik-Artikeln

(57) Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige Folie aus einem oder mehreren biologisch abbaubaren kompostierbaren Polymeren, die in mindestens einer Schicht mindestens ein Absorbens zur Bindung gasförmiger Geruchs- und Aromastoffe enthält.

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Beschreibung

Die Erfindung betrifft eine ein- oder mehrschichtige, biologisch abbaubare, thermoplastische Folie mit verbesserten Barriereeigenschaften gegenüber Gerüchen und Aromen.

Es ist bekannt, daß bestimmte Polymere wie auch andere Werkstoffe einem biologischen Abbau unterliegen können. Hauptsächlich sind hier Materialien zu nennen, die aus natürlich vorkommenden Polymeren direkt oder nach Modifizierung erhalten werden, beispielsweise Polyhydroxyalkanolate wie Polyhydroxybutyrat, plastische Cellulosen, Cellulosester, plastische Stärken, Chitosan und Pullulan. Eine gezielte Variation der Polymerzusammensetzung oder der Strukturen, wie sie von seiten der Polymeranwendung wünschenswert ist, ist bei derartigen Polymeren aufgrund des natürlichen Synthesevorgangs nur schwer und oftmals nur sehr eingeschränkt möglich. Unter den Begriffen "biologisch abbaubare und kompostierbare Polymere bzw. Folien" werden im Sinne dieser Erfindung Güter verstanden, die entsprechend der Prüfung nach DIN V 54 900 von 1998/1999 die "Bioabbaubarkeit" testiert bekommen.

Viele der synthetischen Polymere hingegen werden durch Mikroorganismen nicht oder nur äußerst langsam angegriffen. Hauptsächlich synthetische Polymere, die Heteroatome in der Hauptkette enthalten, werden als potentiell biologisch abbaubar angesehen. Eine wichtige Klasse innerhalb dieser Materialien stellen die Polyester dar. Synthetische Rohstoffe, die nur aliphatische Monomere enthalten, weisen zwar eine relativ gute biologische Abbaubarkeit auf, sind aufgrund ihrer Materialeigenschaften nur äußerst eingeschränkt anwendbar; vergl. Witt et al. in Macrom. Chem. Phys., 195 (1994) S. 793–802. Aromatische Polyester zeigen dagegen bei guten Materialeigenschaften deutlich verschlechterte, biologische Abbaubarkeit.

Aus der DE-A-44 32 161 und der EP-A-641 817 sind in neuerer Zeit verschiedene biologisch abbaubare, synthetische Polymere auf Polyester- bzw. Polyesteramid-Basis bekannt geworden. Diese besitzen die Eigenschaft, daß sie gut thermoplastisch verarbeitbar und auf der anderen Seite biologisch abbaubar sind, d. h. deren gesamte Polymerkette von Mikroorganismen (Bakterien und Pilzen) mittels Enzyme gespalten und vollständig zu Kohlendioxid, Wasser und Biomasse abgebaut werden und ein entsprechender Test in natürlicher Umgebung unter Einwirkung von Mikroorganismen die Bedingungen für Bioabbaubarkeit gemäß DIN V 54 900 erfüllen. Diese biologisch abbaubaren Materialien können aufgrund des thermoplastischen Verhaltens zu Halbzeugen wie Gieß- oder Blasfilmen verarbeitet werden. Dennoch ist der Einsatz dieser Halbzeuge stark begrenzt. Beispielsweise sind die Sperreigenschaften hinsichtlich Wasserdampf und Gasen im Vergleich zu Filmen aus typischen, jedoch nicht biologisch abbaubaren Kunststoffen wie Polyethylen, Polypropylen oder Polyamid sehr schlecht. Gleiches gilt für die Barriereeigenschaften gegenüber Aromen und Gerüchen. Auch die Herstellung mehrschichtiger Folien aus unterschiedlichen biologisch abbaubaren Materialien verbessert die Barriere nur geringfügig.

Aufgabe der vorliegenden Erfindung war es daher, eine Folie aus biologisch abbaubaren Polymeren mit einer verbesserten Barriere gegenüber Gerüchen und Aromen bereitzustellen.

Dieses Ziel wird dadurch erreicht, daß eine ein- oder mehrschichtige, biologisch abbaubare und kompostierbare Folie in mindestens einer Schicht mit mindestens einem Absorbens versehen wird. Das Absorbens bindet in der Folie gasförmige Geruchs- und Aromastoffe. Bei mehrschichtigen Folien können grundsätzlich alle Schichten mit einem Absorbens oder mehreren Absorbentien ausgerüstet werden, bevorzugt sind jedoch Ausführungsformen, in denen die Folie mindestens dreischichtig ist und die beiden äußeren Folienschichten kein Absorbens enthalten. Gegebenenfalls können verschiedene Folienschichten auch unterschiedliche Absorbentien enthalten.

Bevorzugt als Absorbens verwendet werden geruchsabsorbierende Feststoffe, die sich in fein verteilter Form mit möglichst geringen Teilchengrößen herstellen lassen, mit den zur Herstellung der Folie eingesetzten biologisch abbaubaren Polymeren verträglich sind und deren Schmelzpunkt oberhalb den bei der Herstellung der Folie auftretenden Temperaturen liegt. Insbesondere werden als Absorbens Zeolithe eingesetzt, wie sie auf S. 475–501 von Ullmann's Encyclopedia of Industrial Chimistry, 5. Aufl. 1996, VCH Weinheim, beschrieben sind.

Der Gehalt der erfindungsgemäßen Folie an Absorbens beträgt vorzugsweise 0,01 bis 25 Gew.-%, insbesondere 0,01 bis 10 Gew.-%, jeweils bezogen auf das Gesamtgewicht der jeweiligen mindestens ein Absorbens enthaltenden Folienschicht.

Dabei war es überraschend, das Absorbens bzw. die Absorbentien auch bei Einbetten in die Folienmatrix weiterhin eine ausreichende Absorbtionswirkung zeigen und sich auch in größeren Mengen ohne negativen Einfluß auf andere Folieneigenschaften in die erfindungsgemäße Folie einarbeiten lassen.

In einer besonderen Form kann die erfindungsgemäße ein- oder mehrschichtige Folie zusätzlich mit einer Corona- und/oder Flamm- und/oder Plasmavorbehandlung und/oder einem oxidativ wirkenden Stoff und/oder einem an-/ablagerbaren Stoff und/oder einem Stoffgemisch aus oxidativ wirkenden und/oder anlagerbaren Stoffen, vorzugsweise Gasen, mit radikalischen Komponenten wie Ozon oder einem plasmaangeregten Gasgemisch aus beispielsweise Hexamethyldisiloxan mit Stickstoff (N_2) und/oder Sauerstoff (O_2), auf der Oberfläche behandelt werden. Dabei erfolgen die genannten Oberflächenvorbehandlungen bei verstreckten Folien vorzugsweise nach der Orientierung.

Die monoaxiale oder biaxiale Orientierung erfolgt bei amorphen Thermoplasten in Temperaturbereichen oberhalb der Glasübergangstemperatur sowie bei teilkristallinen Thermoplasten oberhalb der Glasübergangstemperatur und unterhalb der Kristallitschmelztemperatur.

Als Polymere für die erfindungsgemäße ein- oder mehrschichtige Folie mit verbesserten Barriereeigenschaften sind geeignet:
aliphatische oder teilaromatische Polyester aus

A) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie vorzugsweise Ethandiol, Butandiol, Hexandiol oder besonders bevorzugt Butandiol und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, bevorzugt mit 5 oder 6 C-Atomen im cycloaliphatischen Ring, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevor-

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zugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, bevorzugt C_3-C_{12}-Alkyldiolen, wie beispielsweise Neopentyglykol, und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen wie beispielsweise 1,2,3-Propantriol oder Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren, vorzugsweise C_2-C_{12}-Alkyldicarbonsäuren, wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder

B) aus säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,

oder einer Mischung und/oder einem Copolymer aus A und B, wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;
aliphatische oder teilaromatische Polyesterurethane aus

C) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie beispielsweise Ethandiol, Butandiol, Hexandiol, besonders bevorzugt Butandiol und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, vorzugsweise mit C_5- oder C_6 cycloaliphatischem Ring, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkyldiolen, wie beispielsweise Neopentyglykol, und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkylpolyolen, wie beispielsweise 1,2,3-Propantriol oder Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren, vorzugsweise C_2-C_{12}-Alkyldicarbonsäuren, wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure, und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder

D) aus säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen, beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,

oder einer Mischung und/oder einem Copolymer aus C und D, wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;
und

E) aliphatischen und/oder cycloaliphatischen bifunktionellen und zusätzlich gegebenenfalls höherfunktionellen Isocyanaten, mit vorzugsweise 1 bis 12 C-Atomen bzw. 5 bis 8 C-Atomen im Falle von cycloaliphatischen Isocyanaten, z. B. Tetramethylendiisocyanat, Hexamethylendiisocyanat, Isophorondiisocyanat, gegebenenfalls zusätzlich mit linearen und/oder verzweigten und/oder cycloaliphatischen bifunktionellen und/oder höherfunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkyldi- oder -polyole bzw. 5 bis 8 C-Atomen im Falle von cycloaliphatischen Alkoholen, z. B. Ethandiol, Hexandiol, Butandiol, Cyclohexandimethanol, und/oder gegebenenfalls zusätzlich mit linearen und/oder verzweigten und/oder cycloaliphatischen bifunktionellen und/oder höherfunktionellen Aminen und/oder Aminoalkoholen mit vorzugsweise 2 bis 12 C-Atomen in der Alkylkette, z. B. Ethylendiamin oder Aminoethanol, und/oder gegebenenfalls weitere modifizierte Amine oder Alkohole wie beispielsweise Ethylendiaminoethansulfonsäure, als freie Säure oder als Salz,

wobei der aus C) und D) gebildete Esteranteil mindestens 75 Gew.-%, bezogen auf die Summe aus C), D) und 13), beträgt;
aliphatische oder aliphatisch-aromatische Polyestercarbonate aus

F) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C2 bis C_{10}-Dialkoholen wie beispielsweise Ethandiol, Butandiol, Hexandiol oder besonders bevorzugt Butandiol und/ oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, vorzugsweise mit 5 bis 8 C-Atomen im cycloaliphatischen Ring, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, vorzugsweise mit C_2-C_{12}-Alkyldicarbonsäuren, wie beispielsweise Neopentyglykol und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen wie beispielsweise 1,2,3-Propantriol, Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder

G) säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette, beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,

oder einer Mischung und/oder einem Copolymer aus F und G, wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;

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und

H) einem Carbonatanteil, der aus aromatischen bifunktionellen Phenolen, bevorzugt Bisphenol-A, und Carbonatspendern, beispielsweise Phosgen, hergestellt wird
oder
einem Carbonatanteil, der aus aliphatischen Kohlensäureestern oder deren Derivaten wie beispielsweise Chlorkohlensäureestern oder aliphatischen Carbonsäuren oder deren Derivaten wie beispielsweise Salzen und Carbonatspendern, beispielsweise Phosgen, hergestellt wird, wobei

der aus F) und/oder G) gebildete Esteranteil mindestens 70 Gew.-%, bezogen auf die Summe aus F), G) und H) beträgt;
aliphatische oder teilaromatische Polyesteramide aus

I) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie beispielsweise Ethandiol, Butandiol, Hexandiol, besonders bevorzugt Butandiol, und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, vorzugsweise mit 5 bis 8 C-Atomen, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, vorzugsweise C_3-C_{12} Alkyldiolen, wie beispielsweise Neopentyglykol und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkylpolyole, wie beispielsweise 1,2,3-Propantriol, Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette, wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder
K) aus säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen in der Kohlenstoffkette, beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,

oder einer Mischung und/oder einem Copolymer aus I) und K), wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;
und

L) aliphatischen und/oder cycloaliphatischen bifunktionellen und/oder gegebenenfalls geringen Mengen verzweigten bifunktionellen Aminen, bevorzugt sind lineare aliphatische C2 bis C_{10}-Diamine, und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Aminen, unter den Aminen bevorzugt Hexamethylendiamin, Isophorondiamin und besonders bevorzugt Hexamethylendiamin, sowie aus linearen und/oder cycloaliphatischen bifunktionellen Säuren, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette bzw. C_5- oder C_6-Ring im Falle von cycloaliphatischen Säuren, bevorzugt Adipinsäure, und/oder ggf. geringen Mengen von verzweigten bifunktionellen und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren, vorzugsweise mit 2 bis 10 C-Atomen, oder
M) säure- und aminfunktionalisierten Bausteinen, vorzugsweise mit 4 bis 20 C-Atomen in der cycloaliphatischen Kette, bevorzugt ω-Laurinlactam, ε-Caprolactam, besonders bevorzugt ε-Caprolactam,

oder einer Mischung aus L) und M), wobei der aus I) und/oder K) gebildete Esteranteil mindestens 20 Gew.-%, bezogen auf die Summe aus I), K), L) und M) beträgt, vorzugsweise der aus I) und/oder K) gebildete Esteranteil 20 bis 85 Gew.-%, und der Anteil der aus L) und/oder M) gebildeten Amidstrukturen 80 bis 15 Gew.-% beträgt.

Bei den in der erfindungsgemäßen Folie verwendeten biologisch abbaubaren Polymeren kann es sich sowohl um reine Polymere als auch um Mischungen aus verschiedenen Polymeren handeln, wobei im Falle von Mischungen vorzugsweise Polymere aus nur einer der vorgenannten Verbindungsklassen (Polyester, Polyesterurethane, Polyestercarbonate, Polyesteramide) verwendet werden.

Die für die erfindungsgemäße ein- oder mehrschichtige Folie eingesetzten Polymere können zusätzlich übliche Additive und Hilfsmittel enthalten.

Bevorzugt kommen in den für die erfindungsgemäße Folie eingesetzten Polymeren folgende Additive und Hilfsmittel zum Einsatz:
Maximal 5 Gew.-%, z. B. 0 bis 5 Gew.-% für Polyester typisch eingesetzte Nukleierungsmittel (beispielsweise 1,5-Naphthalindinatriumsulfonat oder Schichtsilikate, beispielsweise Talkum, oder Keimbildner der Nanoteilchengröße, d. h. mittlerer Teilchendurchmesser < 1 μm, aus beispielsweise Titannitrid, Aluminiumhydroxylhydrat, Bariumsulfat oder Zirkonverbindungen) und/oder maximal 5 Gew.-%, z. B. 0 bis 5 Gew.-% der üblichen Stabilisatoren und Neutralisationsmittel und/oder maximal 5 Gew.-%, z. B. 0 bis 5 Gew.-% der üblichen Gleit- und Trennmittel und/oder maximal 5 Gew.-%, z. B. 0 bis 5 Gew.-% der üblichen Antiblockmittel sowie gegebenenfalls Pigmente zur Einfärbung.

Als Stabilisatoren und Neutralisationsmittel können die üblichen stabilisierend wirkenden Verbindungen für Polyesterverbindungen eingesetzt werden. Deren Zusatzmenge liegt 0 maximal bei 5 Gew.-%.

Besonders geeignet als Stabilisatoren sind phenolische Stabilisatoren, Alkali-/Erdalkalistearate und/oder Alkali-/Erdalkalicarbonate. Phenolische Stabilisatoren werden in einer Menge von 0 bis 3 Gew.-%, insbesondere 0,15 bis 0,3 Gew.-% und mit einer Molmasse von mehr als 500 g/mol bevorzugt. Pentaerythrityl-Tetrakis-3(3,5-di-Tertiärbutyl-4-Hydroxyphenyl)-Propionat oder 1,3,5-Trimethyl-2,4,6-tris(3,5-di-Tertiärbutyl-4-Hydroxybenzyl)-benzol sind besonders vorteilhaft. Neutralisationsmittel sind vorzugsweise Dihydrotalcit, Calciumstearat, Calciumcarbonat und/oder Cal-

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ciummontanat einer mittleren Teilchengröße von höchstens 0,7 µm, einer absoluten Teilchengröße von kleiner 10 µm und einer spezifischen Oberfläche von mindestens 40 m^2/g.

In einer besonders bevorzugten Aufführungsform der Folie besitzt diese einen Nukleierungsmittelanteil von 0,0001 bis 2 Gew.-% und einen Stabilisatoren- und Neutralisationsmittelanteil von 0,0001 bis 2 Gew.-%.

Gleit- und Trennmittel sind höhere aliphatische Amide, tertiäre Amine, aliphatische Säureamide, höhere aliphatische Säureester, niedrigmolekulare polarmodifizierte Wachse, Montanwachse, cyclische Wachse, Phthalate, Metallseifen sowie Silikonöle. Besonders geeignet ist der Zusatz von höheren aliphatischen Säureamiden und Silikonölen.

Unter den aliphatischen Amiden sind insbesondere die Angebotsformen von Ethylenamid bis Stearylamid geeignet. Aliphatische Säureamide sind Amide einer wasserunlöslichen Monocarbonsäure (sogenannte Fettsäuren) mit 8 bis 24 Kohlenstoffatomen, vorzugsweise 10 bis 18 Kohlenstoffatomen. Erucasäureamid, Stearinsäureamid und Ölsäureamid sind hierunter bevorzugt.

Geeignet als Trenn- oder Gleitmittel sind außerdem Verbindungen, die sowohl Ester- als auch Amidgruppen enthalten, wie beispielsweise Stearamid-Ethylstearat bzw. 2 Stear-Amido-Ethyl-Stearat.

Unter der Bezeichnung Montanwachse fällt ein Reihe von verschiedenen Verbindungen. Siehe hierzu Neumüller et al. in Römpps Chemie-Lexikon, Franckh'sche Verlagshandlung, Stuttgart, 1974.

Als cyclische Wachse sind beispielsweise Komponenten wie cyclische Adipinsäuretetramethylenester bzw. 1.6-Dioxa-2.7-dioxocyclododecan, oder das homologe Hexamethylenderivat geeignet. Solche Stoffe sind als Handelsprodukte mit dem Namen Glycolube VL bekannt.

Geeignete Silikonöle sind Polydialkylsiloxane, vorzugsweise Polydimethylsiloxan, Polymethylphenylsiloxan, olefinmodifiziertes Silikon, mit Polyethern modifiziertes Silikon wie z. B. Polyethylenglykol und Polypropylenglykol sowie epoxyamino- und alkoholmodifiziertes Silikon. Die Viskosität der geeigneten Silikonöle liegt im Bereich von 5000 bis 1 000 000 mm^2/s. Polydimenthylsiloxan mit einer Viskosität von 10.000 bis 100.000 mm^2/s ist bevorzugt.

Die Menge des zugesetzten Gleitmittels beträgt maximal 5 Gew.-%. In einer besonders bevorzugten Ausführungsform der Folie besitzt diese einen Gleitmittelanteil von 0,005 bis 4 Gew.-%. In einer ganz besonders bevorzugten Ausführungsform der Folie besitzt diese einen Gleitmittelanteil von 0,05 bis 1 Gew.-%.

Geeignete Antiblockmittel sind sowohl anorganische als auch organische Zusatzstoffe, die aufgrund ihrer Teilchengröße und/oder Form als Erhebung aus der Folienoberfläche herausragen und somit einen Abstandshaltereffekt hervorrufen.

In einer bevorzugten Form werden als anorganische Antiblockmittel die folgenden Stoffe eingesetzt:
Aluminiumhydroxid
Aluminiumsilikate, beispielsweise Kaolin oder Kaolinton,
Aluminiumoxide, beispielsweise Θ-Aluminiumoxid
Aluminiumsulfat
Keramiken aus Silica-Aluminiumoxiden
Bariumsulfat
natürliche und synthetische Kieselsäuren
Schichtsilikate, beispielsweise Asbest,
Siliciumdioxid
Calciumcarbonat vom Calcit-Typ
Calciumphosphat
Magnesiumsilikate
Magnesiumcarbonat
Magnesiumoxid
Titandioxid
Zinkoxid
Microglaskugeln
und als organische Antiblockmittel die folgenden Stoffe eingesetzt:
mit dem biologisch abbaubaren Polymer unverträgliche organische Polymerisate wie Stärke
Polystyrole
Polyamide
Polycarbonate
vernetztes und unvernetztes Polymethylmethacrylat
vernetztes Polysiloxan (z. B. Tospearl)
polarmodifizierts Polyethylen (z. B. Maleinsäureanhydrid-gepfropftes Polyethylen)
polarmodifizierts Polypropylen (z. B. Maleinsäureanhydrid-gepfropftes Polypropylen)
statistische Copolymer auf Ethylen- oder Propylenbasis mit Vinylalcohol oder Vinylacetat oder Acrylsäure oder Acrylsäureester oder Methacrylsäure oder Methacrylsäureester oder Metallsalzen der Methacrylsäure oder Metallsalze der Methacrylsäureester
Benzoguanamin Formaldehyd Polymere
aliphatische und teilaromatische Polyester mit anderen Schmelzpunkten als der Folienrohstoff
aliphatische Polyesteramide mit anderen Schmelzpunkten als der Folienrohstoff
aliphatische Polyesterurethane mit anderen Schmelzpunkten als der Folienrohstoff
aliphatisch-aromatische Polyestercarbonate mit anderen Schmelzpunkten als der Folienrohstoff.

Die wirksame Menge an Antiblockmittel liegt im Bereich von 0 bis maximal 5 Gew.-%. In einer besonders bevorzugten Ausführung enthält die Folie 0,005 bis 4 Gew.-% Antiblockmittel. In einer ganz besonders bevorzugten Ausführung enthält die Folie 0,05 bis 1 Gew.-% Antiblockmittel. Die mittlere Teilchengröße liegt zwischen 1 und 6 µm, insbesondere 2 und 5 µm, wobei Teilchen mit einer kugelförmigen Gestalt, wie in der EP-A-0 236 945 und der DE-A-38 01 535 beschrieben, besonders geeignet sind. Besonders geeignet sind auch Kombinationen verschiedener Abstandshaltersysteme.

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In einer besonders bevorzugten Ausführung werden die Abstandshaltersysteme den äußeren Deckschichten zugegeben.

Die erfindungsgemäße biologisch abbaubare und kompostierbare ein- oder mehrschichtige Folie mit verbesserten Barriereeigenschaften kann Pigmente zur Einfärbung enthalten. Dabei kann es sich um organische oder anorganische Pigmente, die der Deckschicht, der Zwischenschicht oder den Zwischenschichten oder allen Schichten zugegeben werden. In einer besonders bevorzugten Form sind die Pigmente biologisch unbedenklich und/oder haben eine Konzentration von weniger als 1 Gew.-%.

Zur Herstellung der erfindungsgemäßen Folie werden die Polymere für die gegebenenfalls mit Additiven versehene Folie bzw. die einzelnen Folienschichten gemäß dem üblichen Stand der Technik mit den gewünschten Gewichtsmengen an Adsorbens sowie organischen oder anorganischen Additiven und/oder Hilfsstoffen ausgerüstet. Dies geschieht beim Granulieren des Rohstoffs beispielsweise in Zweischneckenextrudern, wo dem Rohstoff die Additive und/oder Absorbentien zugegeben werden. Neben dieser Art der Additivierung und Zugabe der Absorbentien besteht auch die Möglichkeit, daß ein Teil oder alle notwendigen Additive und/oder Absorbentien einem nicht oder teilweise ausgerüsteten Rohstoff in Form eines Masterbatches zugegeben werden. Unter dem Begriff Masterbatch ist im Rahmen der vorliegenden Erfindung eine Stammischung zu verstehen, insbesondere ein granulatförmiges staubfreies Konzentrat eines Kunststoffrohstoffes mit hohen Mengen an Additiven und/oder Absorbentien, das in der Masseaufbereitung als Zwischenprodukt verwendet wird (als Materialzusatz zu einem nicht oder nur teilweise oder unvollständig mit Additiven und/oder Absorbentien ausgerüstetem Granulat), um daraus Folien herzustellen, die eine bestimmte Menge an Additiven enthalten. Das Masterbatch wird vor dem Einfüllen des Polymergranulates in den Extruder in derartigen Mengen zu den nicht oder nur teilweise oder unvollständig mit Additiven und/oder Absorbentien ausgerüsteten Rohstoffen zugemischt, so daß die gewünschten Gewichtsprozentanteile an Additiven und/oder Absorbentien in den Folien realisiert werden. Die Materialien, aus denen neben den Additiven und/oder Absorbentien die Masterbatche hergestellt werden, sind in einer besonders bevorzugten Form ebenfalls biologisch abbaubar, biologisch unbedenklich, oder haben einen Gewichtsanteil, der kleiner als 1 Gew.-% ist.

Die bevorzugten Materialien, aus denen neben den Additiven die Masterbatche hergestellt werden, sind Stoffe, die mit den in dieser Erfindung genannten Polymeren verträglich sind.

Die Herstellung der erfindungsgemäßen ein- oder mehrschichtigen Folie geschieht zweckmäßig über ein Extrusionsverfahren. Die in Granulatform, gegebenenfalls einschließlich der gewünschten Additive und Hilfsstoffe, vorliegenden Polymere werden in einem oder mehreren Extrudern aufgeschmolzen, homogenisiert, komprimiert und über eine Ein- oder Mehrschichtdüse ausgetragen. Dabei kann es sich bei der Düse um eine Ringdüse zur Herstellung einer nahtlosen Schlauchfolie oder eine Flachdüse zur Herstellung einer Flachfolie handeln. Die so ausgetragene bzw. z. B. mittels Walzenpresseuren ausgezogene Folie wird anschließend bis zur Verfestigung abgekühlt. Die Kühlung kann dabei sowohl über Luft als auch über Wasser oder auch mittels Kühlwalzen erfolgen. Die Kühlung kann dabei einseitig oder beidseitig, im Falle einer Schlauchfolie innen- und außenseitig oder nur innen- oder nue außenseitig erfolgen. Die Schlauchfolie kann zudem einseitig oder beidseitig geschnitten werden, so daß man eine mehrschichtige Flachfolie erhält.

Die Folie kann weiterhin nach der Abkühlung bei teilkristallinen Werkstoffen unter Kristallitschmelztemperatur und bei amorphen Werkstoffen oberhalb der Glasübergangstemperatur temperiert und dann ein oder mehrfach monoaxial oder biaxial verstreckt werden, wobei das biaxiale Verstrecken nacheinander oder, insbesondere im Fall von Schlauchfolien, simultan erfolgen kann. Nach der oder den Verstreckstufen kann jeweils optional eine Fixierung der Folie durch eine Wärmebehandlung erfolgen. Ein besonders geeignetes Verstreckverfahren ist dabei für das simultane biaxiale Recken von Schlauchfolien die sogenannte Double-Bubble-Technologie, bei der die Verstreckung einer Primärblase über einen anliegenden Innendruck erfolgt. Zur gezielten Einstellung von Schrumpfeigenschaften kann die Folie anschließend einer Wärmebehandlung unterzogen werden. Die Folie kann in diesem Fixierprozess wieder bis kurz unter die Kristallitschmelztemperatur erwärmt werden.

Sofern anschließend eine Coronabehandlung erfolgen soll, wird zwecksmäßigerweise so vorgegangen, daß die Folie zwischen zwei als Elektroden dienenden Leiterelementen hindurchgeführt wird, wobei zwischen den Elektroden eine so hohe Spannung, meist Wechselspannung (etwa 5 bis 20 kV und 5 bis 30 kHz), angelegt ist, daß Sprüh- oder Coronaentladungen stattfinden können. Durch die Sprüh- oder Coronaentladung wird die Luft oberhalb der Folienoberfläche ionisiert und reagiert mit den Molekülen der Folienoberfläche, so daß zusätzlich polare Einlagerungen in der Polymermatrix entstehen.

Für eine gegebenenfalls an den Reck- bzw. Fixierprozeß anschließende Flammbehandlung mit polarisierter Flamme (vgl. US-A-4,622,237) wird eine elektrische Gleichspannung zwischen einem Brenner (negativer Pol) und einer Kühlwalze angelegt. Die Höhe der angelegten Spannung beträgt zwischen 400 und 3000 V, vorzugsweise liegt sie im Bereich von 500 bis 2000 V. Durch die angelegte Spannung erhalten die ionisierten Atome eine erhöhte Beschleunigung und treffen mit größerer kinetischer Energie auf die Polymeroberfläche. Die chemischen Bindungen innerhalb des Polymermoleküls werden leichter aufgebrochen und die Radikalbildung geht schneller vonstatten. Die thermische Belastung des Polymeren ist hierbei weitaus geringer als bei der Standardflammbehandlung, und es können Folien erhalten werden, bei denen die Siegeleigenschaften der behandelten Seite sogar besser sind als diejenigen der nicht behandelten Seite.

Bei einer sich gegebenenfalls an den Reck- bzw. Fixierprozeß anschließende Plasmavorbehandlung werde in einer Niederdruckkammer Gase, z. B. Sauerstoff oder Stickstoff oder Kohlendioxid oder Methan oder Halogenkohlenwasserstoffe oder Silanverbindungen oder höhermolekulare Verbindungen oder auch Mischungen hiervon, einem hochenergiereichen Feld, z. B. Mikrowellenstrahlung, ausgesetzt. Es entstehen hochenergiereiche Elektronen, die auf die Moleküle treffen und ihre Energien übertragen. Hierdurch entstehen lokal radikalische Strukturen, deren Anregungszustände Temperaturen von einigen zehntausend Grad Celsius entsprechen, obwohl das Plasma selbst sich nahezu auf Raumtemperatur befindet. Dadurch besteht die Möglichkeit, chemische Bindungen aufzubrechen und Reaktionen in Gang zu setzen, die normalerweise erst bei hohen Temperaturen ablaufen können. Es bilden sich Monomerradikale und Ionen. Aus den entstandenen Monomerradikalen bilden sich – teilweise schon im Plasma – kurzkettige Oligomere, die dann auf der zu beschichtenden Oberfläche kondensieren und polymerisieren. Auf dem Beschichtungsgut scheidet sich dadurch ein homogener Film ab.

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Bevor sich jedoch die Monomerradikale bzw. Ionen auf dem Substrat abscheiden, besteht außerdem zusätzlich die Möglichkeit, in der sogenannten After-Glow-Zone einen weiteren Stoffstrom den angeregten Molekülen hinzuzufügen. Hierdurch ist es möglich, einen Stoff oder ein Stoffgemisch zu erzeugen, das bei dem Auftreffen auf der Polymerfilmoberfläche einen oxidativen Angriff bei den Substratpolymeren erzeugt. Es bildet sich eine glasartige und meist hochvernetzte Schicht, die mit der Folienoberfläche fest verbunden ist. Bei geeigneter Stoffzusammensetzung ergibt sich hierdurch einen Anstieg der Oberflächenspannung auf der Folie.

Zudem können bei mehrschichtigen Ausführungsformen der erfindungsgemäßen Folie weitere Adsorbens enthaltene oder Adsorbens-freie Folienschichten durch einen Kaschierprozeß, bei dem die Kaschierfolie als Zulauf durch Naß- oder Thermokaschierung auf die ursprüngliche Folie aufgebracht wird, hergestellt werden.

Bei der Naßkaschierung wird die ursprüngliche Folie oder die Kaschierfolie oder sowohl ursprüngliche Folie als auch Kaschierfolie mit einem Kaschierkleber benetzt, beide Folien durch Walzenpresseure miteinander verbunden und anschließend getrocknet. Als Kaschierkleber kommen sowohl lösungsmittelfreie als auch lösungsmittelhaltige Kleber in Betracht. In einer besonders bevorzugten Form ist der Kaschierkleber ebenfalls biologisch abbaubar und kompostierbar.

Bei der Thermokaschierung wird als zulaufende Kaschierfolie eine Folie verwendet, die durch temperierte Walzenpresseure auf die beschichtete Folie gepreßt wird und damit mit der beschichtete Folienseite verbunden wird. In einer besonders bevorzugten Form wird als zulaufende Kaschierfolie eine mehrschichtige Folie verwendet, die aus mindestens zwei Schichten bestehen, deren Schmelzpunkte verschieden sind. Durch Temperieren der Kaschierfolie auf die Schmelz- oder Erweichungstemperatur der früher schmelzenden Schicht und Anpressen auf die ursprüngliche Folienseite wird die Verbindung zwischen beschichteter Folie und Kaschierfolie erreicht, wobei die früher schmelzende Schicht der Kaschierfolie der ursprünglichen Seite zugewandt ist.

Die Kaschierfolie, die durch Naß- oder Thermokaschierung mit der ursprünglichen Folie verbunden wird, kann sowohl ungereckt, monoaxial als auch biaxial gereckt sein.

Gegenstand der Erfindung ist darüber hinaus die Anwendung der erfindungsgemäßen Folie. Als bevorzugte Anwendung kommt der Einsatz dieser Folie insbesondere als mehrschichtige Folie in vorbehandelter oder unvorbehandelter sowie in bedruckter, unbedruckter oder lackierter Form für die Verpackung insbesondere von geruchsintensiven Lebensmitteln oder Nichtlebensmitteln oder für Schutz- und Trennfunktionen bei Kosmetik und Hygieneartikeln, beispielsweise für Babywindeln oder Damenbinden in Betracht.

Die folgenden Beispiele sollen die Erfindung weiter erläutern, ohne sie darauf zu beschränken:

Beispiele

Beispiel 1 (Vergleichsbeispiel)

Aus zwei unterschiedlichen biologisch abbaubaren und kompostierbaren Polymeren wurde an einer Drei-Schicht-Blasfolienanlage ein symmetrischer, coextrudierter Film mit einem ABA-Aufbau hergestellt. Das Material A Polyesteramid aus 58 Gew.-% Caprolactam, 23 Gew.-% Adipinsäure, 19% Butandiol, (LP BAK 403-004, Bayer AG) hat einen MFI von 9 (in g/10 min bei 190°C, 2,16 kg, gemessen nach DIN 53 735), einen Schmelzpunkt von 125°C, gemessen nach ISO 3146/C2, einen Anteil an Gleitmittel von 0,15 Gew.-% und einen Antiblockanteil bzw. Nukleierungsmittelgehalt von 0,1 Gew.-%. Das Material B Polyesteramid aus 62 Gew.-% Caprolactam, 21 Gew.-% Adipinsäure, 17 Gew.-% Butandiol (LP BAK 404-002, Bayer AG) hat einen MFI von 6 (in g/10min bei 190°C, 2,16 kg, gemessen nach DIN 53 735), einen Schmelzpunkt von 140°C, gemessen nach ISO 3146/C2 und einen Antiblockanteil bzw. Nukleierungsmittelgehalt von 0,1 Gew.-%. Die maximale Extrusionstemperatur betrug 190°C. Die Schmelze wurde über eine Mehrschichtringdüse ausgetragen und durch Luft abgekühlt. Die maximale Düsentemperatur betrug 185°C. Es wurde in diesem Fall eine Folie mit einer Gesamtdicke von 80 µm in einer Breite von 700 mm hergestellt, wobei das Material A mit dem niedrigen Schmelzpunkt die Außenschichten bildet.

Beispiel 2

Aus den gleichen biologisch abbaubaren Polymeren A und B aus Beispiel 1 wurde an der gleichen Blasfolienanlage unter Berücksichtigung der selben Prozeßparametern ein symetrischer ABA-Film hergestellt. Im Gegensatz zum Beispiel 1 wurde dem Material B der innenliegenden Kernschicht 20 Gew.-% eines Masterbatches zugegeben, das 18,4 Gew.-% des Absorbers Mordenit Natrium-Form aktiv (Hersteller: Bayer AG) enthält. Es wurde ein Film mit einer Dicke von 75 µm hergestellt.

Beispiel 3

Aus den biologisch abbaubaren Polymeren A und B aus Beispiel 1 wurde an der gleichen Blasfolienanlage unter Berücksichtigung der selben Prozeßparametern ein symetrischer ABA-Film hergestellt. Im Gegensatz zum Beispiel 1 und 2 wurde dem Material B der innenliegenden Kernschicht 20 Gew.-% eines Masterbatches zugegeben, das 18,8 Gew.-% des Absorbers Zeolith-X Natrium-Form aktiv (Hersteller: Bayer AG) enthält. Es wurde ein Film mit einer Dicke von 80 µm hergestellt.

Beispiel 4

Aus den biologisch abbaubaren Polymeren A und B aus Beispiel 1 wurde an der gleichen Blasfolienanlage unter Berücksichtigung der selben Prozeßparametern ein symetrischer ABA-Film hergestellt. Im Gegensatz zum Beispiel 1, 2 und 3 wurde dem Material B der innenliegenden Kernschicht 20 Gew.-% eines Masterbatches zugegeben, das 19,4 Gew.-% des Absorbers Zeolith-A Calcium-, Natrium-Form aktiv (Hersteller: Bayer AG) enthält. Es wurde ein Film

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mit einer Dicke von 80 µm hergestellt.

An den gefertigten Mustern wurden die folgenden physikalischen Eigenschaften wie folgt gemessen:

Mechanische Eigenschaften

An den Proben wurden die mechanischen Größen Reißfestigkeit und Reißdehnung sowohl in Längs- als auch in Querrichtung gemäß DIN 53 455 bestimmt. Der E-Modul in Längs- und Querrichtung wurde entsprechend der DIN 53 457 gemessen. Die Dicke der einzelnen Muster wurde nach DIN 53 370 bestimmt.

Permeationseigenschaften

An den Proben wurde die Durchlässigkeit von Sauerstoff bei einer Temperatur von 23°C und einer relativen Feuchte von 0% und 75% nach DIN 53 380 gemessen. Weiterhin wurde die Durchlässigkeit von Wasserdampf bei einer Temperatur von 23°C und einer relativen Feuchte von 85% gemäß DIN 53 122 bestimmt.

Sensorik-Test

Ein Becherglas wird mit frischen kleingehackten Zwiebel gefüllt, Folien nach Beispiel 1 bis 4 werden so über ein Becherglas gelegt, daß sie die Öffnung vollständig abschließen.

Mittels sensorischer Geruchsprobe wird der Unterschied in der Gasdurchlässigkeit bestimmt.

Folie nach Beispiel 1:

Sofort ist der Geruch der Zwiebeln durch die Folie zu riechen.

Folien nach Beispiel 2–4:

Der Geruch der feingehackten Zwiebeln ist nicht wahrzunehmen, d. h. der Absorber nimmt die Geruchsbestandteile auf und läßt sie nicht durch die Folie.

Siegelnahtfestigkeiten

An den Proben wurde die Festigkeit von Siegelnähten, die an einem Siegelgerät mit beidseitig beheiztem Siegelwerkzeug gefertigt wurden, bestimmt. Unter der Siegelfestigkeit wird die Höchstkraft verstanden, die erforderlich ist, um eine unter definierten Bedingungen (Druck, Zeit, Temperatur) hergestellte Siegelnaht zu trennen. Die Siegelnaht wird in N angegeben, die Streifenbreite als Index hinzugefügt (N/15 mm). Der Siegelbereich ist der Temperaturbereich, in dem eine meßbare Siegelfestigkeit vorhanden ist.

Aus der zu prüfenden Folienbahn wurden zwei fehlerfreie, saubere Probenstreifen entnommen. Zur Siegelung wurden sie mit den zu versiegelnden Flächen aufeinandergelegt und so zwischen die Siegelbacken gehalten, daß die Probe an jeder Seite mindestens 1 cm überstand. Die Siegelung muß senkrecht zur Folienlaufrichtung erfolgen.

Die Siegelfestigkeitsprüfung erfolgte nach folgender Standardbedingung:

Druck: 50 N/cm^2

Zeit: 0,5 sec

Temperatur: in 10 K-Schritten ab 150°C.

Aus der hergestellten Siegelnaht wurde aus der Mitte exakt in Folienlaufrichtung mit einem Prüfstreifenschneider ein 15 mm breiter Prüfstreifen geschnitten. Mit einer Zugfestigkeitsmaschine wurde er auf Siegelfestigkeit untersucht, indem er senkrecht zur Siegelnaht getrennt wurde. Angegeben wurde jeweils der Maximalwert der beim Zerreißen auftretenden Kraft.

Die Ergebnisse der Untersuchungen an den Proben aus Beispiel 1, 2, 3 und 4 sind in der Tabelle aufgeführt.

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	Beispiel 1	Beispiel 2	Beispiel 3	Beispiel 4
Mechanische Eigenschaften				
Dicke [µm]	80	75	80	80
E-Modul längs [MPa]	213	218	232	227
E-Modul quer [Mpa]	303	290	356	330
Reißfestigkeit längs [MPa]	35	34	33	31
Reißfestigkeit quer [MPa]	24	22	18	20
Reißdehnung längs [%]	406	437	363	368
Reißdehnung quer [%]	639	563	522	545
Permeation				
Sauerstoff 23 °C/0 % r. F. [cm³/m²/d/bar]	247	231	234	204
Sauerstoff 23 °C/75 % r. F. [cm³/m²/d/bar]	341	365	355	306
Wasserdampf 23 °C/85 % r. F. [g/m2/d]	120	128	145	130
Sensorik-Test				
wahrnehmbarer Zwiebelgeruch	ja	nein	nein	nein
Siegelfestigkeit				
Notwendige Zerreißkraft [N/15mm]	29 (bei 160°C)	31 (bei 160°C)	25 (bei 160°C)	25 (bei 160°C)
Biologisch abbaubar				
gemäß DIN V 54900	ja	ja	ja	ja

Patentansprüche

1. Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige Folie aus einem oder mehreren biologisch abbaubaren und kompostierbaren Polymeren und üblichen Additiven und/oder Hilfsstoffen, **dadurch gekennzeich-**

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net, daß die Folie in mindestens einer Schicht mindestens ein Absorbens zur Bindung gasförmiger Geruchs- oder Aromastoffe enthält.

2. Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige Folie gemäß Anspruch 1, dadurch gekennzeichnet, daß als Absorbens geruchsabsorbierende Feststoffe, insbesondere Zeolithe, eingesetzt werden.

3. Biologisch abbaubare und kompostierbare, ein oder mehrschichtige Folie gemäß Anspruch 1 oder 2, dadurch gekennzeichnet, daß der Gehalt an Absorbens in der mindestens ein Absorbens enthaltenden Folienschicht oder den mindestens ein Absorbens enthaltenen Folienschichten 0,01 bis 25 Gew.-%, insbesondere 0,01 bis 10 Gew.-%, bezogen auf das Gesamtgewicht der jeweiligen Folienschicht, beträgt.

4. Biologisch abbaubare und kompostierbare Folie gemäß einem der Ansprüchen 1 bis 3, dadurch gekennzeichnet, daß die Folie mindestens dreischichtig ist und die beiden äußeren Folienschichten kein Absorbens enthalten.

5. Biologisch abbaubare und kompostierbare Folie gemäß einem der Ansprüche 1 bis 4, dadurch gekennzeichnet, daß die Folie mehrschichtig ist und in mindestens zwei Schichten unterschiedliche Absorbentien enthält.

6. Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige Folie gemäß einem der Ansprüche 1 bis 5, dadurch gekennzeichnet, daß die Folie ungereckt oder monoaxial gereckt oder biaxial gereckt ist.

7. Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige Folie gemäß einem der Ansprüche 1 bis 6, dadurch gekennzeichnet, dass es die biologisch abbaubaren und kompostierbaren Polymere, Polyesterpolymere aus der Gruppe Polyester, Polyesterurethane, Polyestercarbonate, Polyesteramide sind.

8. Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige Folie gemäß einem der Ansprüche 1 bis 7, dadurch gekennzeichnet, daß es sich bei den biologisch abbaubaren und kompostierbaren Polymeren um

aliphatische oder teilaromatische Polyester aus

A) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie beispielsweise Ethandiol, Butandiol, Hexandiol oder besonders bevorzugt Butandiol und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, bevorzugt mit 5 oder 6 C-Atomen im cycloaliphatischen Ring, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, bevorzugt C_3-C_{12}-Alkyldiolen, wie beispielsweise Neopentyglykol, und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen wie beispielsweise 1,2,3-Propantriol oder Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren, vorzugsweise C_2-C_{12} Alkyldicarbonsäuren, wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder

B) aus säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,

oder einer Mischung und/oder einem Copolymer aus A und B, wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;

aliphatische oder teilaromatische Polyesterurethane aus

C) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie beispielsweise Ethandiol, Butandiol, Hexandiol, besonders bevorzugt Butandiol und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, vorzugsweise mit C_5- oder C_6-cycloaliphatischem Ring, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkyldiolen, wie beispielsweise Neopentyglykol, und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkylpolyolen, wie beispielsweise 1,2,3-Propantriol oder Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren, vorzugsweise C_2-C_{12}-Alkyldicarbonsäuren, wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure, und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder

D) aus säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen, beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,

oder einer Mischung und/oder einem Copolymer aus C und D, wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;

und

E) aliphatischen und/oder cycloaliphatischen bifunktionellen und zusätzlich gegebenenfalls höherfunktionellen Isocyanaten, mit vorzugsweise 1 bis 12 C-Atomen bzw. 5 bis 8 C-Atomen im Falle von cycloaliphatischen Isocyanaten, z. B. Tetramethylendiisocyanat, Hexamethylendiisocyanat, Isophorondiisocyanat, gegebenenfalls zusätzlich mit linearen und/oder verzweigten und/oder cycloaliphatischen bifunktionellen und/oder höherfunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkyldi- oder -polyole bzw. 5 bis 8 C-Atomen im Falle von cycloaliphatischen Alkoholen, z. B. Ethandiol, Hexandiol, Butandiol, Cyclohexandimethanol, und/oder gegebenenfalls zusätzlich mit linearen und/oder verzweigten und/oder cycloaliphatischen bifunktionellen und/oder höherfunktionellen Aminen und/oder Aminoalkoholen mit vorzugsweise 2 bis 12 C-Atomen in der Alkylkette, z. B. Ethylendiamin oder Aminoethanol, und/oder gegebenenfalls weitere modifizierte Amine oder Alkohole wie beispielsweise Ethylendiaminoethansulfonsäure, als freie Säure oder als Salz,

wobei der aus C) und/oder D) gebildete Esteranteil mindestens 75 Gew.-%, bezogen auf die Summe aus C), D) und

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E), beträgt;
aliphatische oder aliphatisch-aromatische Polyestercarbonate aus
F) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie beispielsweise Ethandiol, Butandiol, Hexandiol oder besonders bevorzugt Butandiol und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, vorzugsweise mit 5 bis 8 C-Atomen im cycloaliphatischen Ring, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, vorzugsweise mit C_2-C_{12}-Alkyldicarbonsäuren, wie beispielsweise Neopentyglykol und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen wie beispielsweise 1,2,3-Propantriol, Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder
G) säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette, beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,
oder einer Mischung und/oder einem Copolymer aus F und G, wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;
und
H) einem Carbonatanteil, der aus aromatischen bifunktionellen Phenolen, bevorzugt Bisphenol-A, und Carbonatspendern, beispielsweise Phosgen, hergestellt wird
oder
einem Carbonatanteil, der aus aliphatischen Kohlensäureestern oder deren Derivaten wie beispielsweise Chlorkohlensäureestern oder aliphatischen Carbonsäuren oder deren Derivaten wie beispielsweise Salzen und Carbonatspendern, beispielsweise Phosgen, hergestellt wird, wobei
der aus F) und /oder G) gebildete Esteranteil mindestens 70 Gew.-%, bezogen auf die Summe aus F), G) und H) beträgt;
aliphatische oder teilaromatische Polyesteramide aus
I) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie beispielsweise Ethandiol, Butandiol, Hexandiol, besonders bevorzugt Butandiol, und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, vorzugsweise mit 5 bis 8 C-Atomen, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkyldiolen, wie beispielsweise Neopentyglykol und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkylpolyole, wie beispielsweise 1,2,3-Propantriol, Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette, wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder
K) aus säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen in der Kohlenstoffkette, beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,
oder einer Mischung und/oder einem Copolymer aus I) und K), wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;
und
L) aliphatischen und/oder cycloaliphatischen bifunktionellen und/oder gegebenenfalls geringen Mengen verzweigten bifunktionellen Aminen, bevorzugt sind lineare aliphatische C_2 bis C_{10}-Diamine, und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Aminen, unter den Aminen bevorzugt Hexamethylendiamin, Isophorondiamin und besonders bevorzugt Hexamethylendiamin, sowie aus linearen und/oder cycloaliphatischen bifunktionellen Säuren, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette bzw. C_5- oder C_6-Ring im Falle von cycloaliphatischen Säuren, bevorzugt Adipinsäure, und/oder ggf. geringen Mengen von verzweigten bifunktionellen und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren, vorzugsweise mit 2 bis 10 C-Atomen, oder
M) säure- und aminfunktionalisierten Bausteinen, vorzugsweise mit 4 bis 20 C-Atomen in der cycloaliphatischen Kette, bevorzugt ω-Laurinlactam, ε-Caprolactam, besonders bevorzugt ε-Caprolactam,
oder einer Mischung aus L) und M), wobei der aus I) und/oder K) gebildete Esteranteil mindestens 20 Gew.-%, bezogen auf die Summe aus I), K), L,) und M) beträgt, vorzugsweise der aus I) und/oder K) gebildete Esteranteil 20 bis 85 Gew.-% und der Anteil der aus L) und/oder M) gebildeten Amidstrukturen 80 bis 15 Gew.-% beträgt, handelt.

9. Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige Folie gemäß einem der Ansprüche 1 bis 8, dadurch gekennzeichnet, daß es sich bei den erfindungsgemäßen biologisch abbaubaren und kompostierbaren Polymeren sowohl um reine Polymere als auch um Mischungen aus verschiedenen Polymeren handeln kann.

10. Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige gemäß einem der Ansprüche 1 bis 9, dadurch gekennzeichnet, daß als Additive und/oder Hilfsmittel 0 bis maximal 5 Gew.-% Nukleierungsmitteln und 0

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bis maximal 5 Gew.-% der üblichen Stabilisatoren und Neutralisationsmittel und 0 bis maximal 5 Gew.-% der üblichen Gleit- und Trennmittel und Pigmente zur Einfärbung der Folie in einer, mehreren oder allen Schichten sowie 0 bis maximal 5 Gew.-% der üblichen Antiblockmittel bevorzugt in einer oder den äußeren Deckschichten enthält, wobei sich die Konzentrationsangaben auf die Gesamtmasse der Folie beziehen.

11. Verwendung der biologisch abbaubaren und kompostierbaren, ein- oder mehrschichtigen Folie gemäß einem der Ansprüche 1 bis 10 in vorbehandelter oder unvorbehandelter sowie in bedruckter, unbedruckter oder lackierter Form für die Verpackung von Lebensmitteln oder Wasserdampf oder Sauerstoff-empfindlichen oder geruchsintensiven Nichtlebensmitteln.

12. Verwendung der biologisch abbaubaren und kompostierbaren, ein- oder mehrschichtigen Folie gemäß einem der Ansprüche 1 bis 10 in Kosmetik oder Hygieneartikeln.

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(12) NACH DEM VERTRAG ÜBER DIE INTERNATIONALE ZUSAMMENARBEIT AUF DEM GEBIET DES PATENTWESENS (PCT) VERÖFFENTLICHTE INTERNATIONALE ANMELDUNG

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— *Mit internationalem Recherchenbericht.*

Zur Erklärung der Zweibuchstaben-Codes, und der anderen Abkürzungen wird auf die Erklärungen ("Guidance Notes on Codes and Abbreviations") am Anfang jeder regulären Ausgabe der PCT-Gazette verwiesen.

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(54) Title: SINGLE OR MULTI-LAYER BIOLOGICALLY DEGRADABLE THERMOPLASTIC FILM

(54) Bezeichnung: EIN- ODER MEHRSCHICHTIGE, BIOLOGISCH ABBAUBARE, THERMOPLASTISCHE FOLIE

(57) Abstract: The invention relates to a biologically degradable and compostable single or multi-layer film comprised of one or more biologically degradable and compostable polymers which, in at least one layer, contain at least one absorbent for binding gaseous odorous substances and aromatic substances.

(57) Zusammenfassung: Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige Folie aus einem oder mehreren biologisch abbaubaren und kompostierbaren Polymeren, die in mindestens einer Schicht mindestens ein Absorbens zur Bindung gasförmiger Geruchs- und Aromastoffe enthält.

Ein- oder mehrschichtige, biologisch abbaubare, thermoplastische Folie

Die Erfindung betrifft eine ein- oder mehrschichtige, biologisch abbaubare, thermoplastische Folie mit verbesserten Barriereeigenschaften gegenüber Gerüchen und Aromen.

Es ist bekannt, daß bestimmte Polymere wie auch andere Werkstoffe einem biologischen Abbau unterliegen können. Hauptsächlich sind hier Materialien zu nennen, die aus natürlich vorkommenden Polymeren direkt oder nach Modifzierung erhalten werden, beispielsweise Polyhydroxyalkanolate wie Polyhydroxybutyrat, plastische Cellulosen, Celluloseester, plastische Stärken, Chitosan und Pullulan. Eine gezielte Variation der Polymerzusammensetzung oder der Strukturen, wie sie von seiten der Polymeranwendung wünschenswert ist, ist bei derartigen Polymeren aufgrund des natürlichen Synthesevorgangs nur schwer und oftmals nur sehr eingeschränkt möglich. Unter den Begriffen "biologisch abbaubare und kompostierbare Polymere bzw. Folien" werden im Sinne dieser Erfindung Güter verstanden, die entsprechend der Prüfung nach DIN V 54 900 von 1998/1999 die "Bioabbaubarkeit" testiert bekommen.

Viele der synthetischen Polymere hingegen werden durch Mikroorganismen nicht oder nur äußerst langsam angegriffen. Hauptsächlich synthetische Polymere, die Heteroatome in der Hauptkette enthalten, werden als potentiell biologisch abbaubar angesehen. Eine wichtige Klasse innerhalb dieser Materialien stellen die Polyester dar. Synthetische Rohstoffe, die nur aliphatische Monomere enthalten, weisen zwar eine relativ gute biologische Abbaubarkeit auf, sind aufgrund ihrer Materialeigenschaften nur äußerst eingeschränkt anwendbar; vergl. Witt et al. in Macrom. Chem. Phys., 195 (1994) S. 793 - 802. Aromatische Polyester zeigen dagegen bei guten Materialeigenschaften deutlich verschlechterte, biologische Abbaubarkeit.

Aus der DE-A-44 32 161 und der EP-A-641 817 sind in neuerer Zeit verschiedene biologisch abbaubare, synthetische Polymere auf Polyester- bzw. Polyesteramid-Basis bekannt geworden. Diese besitzen die Eigenschaft, daß sie gut thermoplastisch verarbeitbar und auf der anderen Seite biologisch abbaubar sind, d.h. deren gesamte Polymerkette von Mikroorganismen (Bakterien und Pilzen) mittels Enzyme gespalten und vollständig zu Kohlendioxid, Wasser und Biomasse abgebaut werden und ein entsprechender Test in natürlicher Umgebung unter Einwirkung von Mikroorganismen die Bedingungen für Bioabbaubarkeit gemäß DIN V 54 900 erfüllen. Diese biologisch abbaubaren Materialien können aufgrund des thermoplastischen Verhaltens zu Halbzeugen wie Gieß- oder Blasfilmen verarbeitet werden. Dennoch ist der Einsatz dieser Halbzeuge stark begrenzt. Beispielsweise sind die Sperreigenschaften hinsichtlich Wasserdampf und Gasen im Vergleich zu Filmen aus typischen, jedoch nicht biologisch abbaubaren Kunststoffen wie Polyethylen, Polypropylen oder Polyamid sehr schlecht. Gleiches gilt für die Barriereeigenschaften gegenüber Aromen und Gerüchen. Auch die Herstellung mehrschichtiger Folien aus unterschiedlichen biologisch abbaubaren Materialien verbessert die Barriere nur geringfügig.

Aufgabe der vorliegenden Erfindung war es daher, eine Folie aus biologisch abbaubaren Polymeren mit einer verbesserten Barriere gegenüber Gerüchen und Aromen bereitzustellen.

Dieses Ziel wird dadurch erreicht, daß eine ein- oder mehrschichtige, biologisch abbaubare und kompostierbare Folie in mindestens einer Schicht mit mindestens einem Absorbens versehen wird. Das Absorbens bindet in der Folie gasförmige Geruchs- und Aromastoffe. Bei mehrschichtigen Folien können grundsätzlich alle Schichten mit einem Absorbens oder mehreren Absorbentien ausgerüstet werden, bevorzugt sind jedoch Ausführungsformen, in denen die Folie mindestens dreischichtig ist und die beiden äußeren Folienschichten kein Absorbens enthalten. Gegebenenfalls können verschiedene Folienschichten auch unterschiedliche Absorbentien enthalten.

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Bevorzugt als Absorbens verwendet werden geruchsabsorbierende Feststoffe, die sich in fein verteilter Form mit möglichst geringen Teilchengrößen herstellen lassen, mit den zur Herstellung der Folie eingesetzten biologisch abbaubaren Polymeren veträglich sind und deren Schmelzpunkt oberhalb den bei der Herstellung der Folie auftretenden Temperaturen liegt. Insbesondere werden als Absorbens Zeolithe eingesetzt, wie sie auf S. 475-501 von Ullmann's Encyclopedia of Industrial Chimistry, 5. Aufl. 1996, VCH Weinheim, beschrieben sind.

Der Gehalt der erfindungsgemäßen Folie an Absorbens beträgt vorzugsweise 0,01 bis 25 Gew.-%, insbesondere 0,01 bis 10 Gew.-%, jeweils bezogen auf das Gesamtgewicht der jeweiligen mindestens ein Absorbens enthaltenden Folienschicht.

Dabei war es überraschend, das Absorbens bzw. die Absorbentien auch bei Einbetten in die Folienmatrix weiterhin eine ausreichende Absorbtionswirkung zeigen und sich auch in größeren Mengen ohne negativen Einfluß auf andere Folieneigenschaften in die erfindungsgemäße Folie einarbeiten lassen.

In einer besonderen Form kann die erfindungsgemäße ein- oder mehrschichtige Folie zusätzlich mit einer Corona- und/oder Flamm- und/oder Plasmavorbehandlung und/oder einem oxidativ wirkenden Stoff und/oder einem an-/ablagerbaren Stoff und/oder einem Stoffgemisch aus oxidativ wirkenden und/oder anlagerbaren Stoffen, vorzugsweise Gasen, mit radikalischen Komponenten wie Ozon oder einem plasmaangeregten Gasgemisch aus beispielsweise Hexamethyldisiloxan mit Stickstoff (N_2) und/oder Sauerstoff (O_2), auf der Oberfläche behandelt werden. Dabei erfolgen die genannten Oberflächenvorbehandlungen bei verstreckten Folien vorzugsweise nach der Orientierung..

Die monoaxiale oder biaxiale Orientierung erfolgt bei amorphen Thermoplasten in Temperaturbereichen oberhalb der Glasübergangstemperatur sowie bei teilkristalli-

nen Thermoplasten oberhalb der Glasübergangstemperatur und unterhalb der Kristallitschmelztemperatur.

Als Polymere für die erfindungsgemäße ein- oder mehrschichtige Folie mit verbesserten Barriereeigenschaften sind geeignet:

aliphatische oder teilaromatische Polyester aus

E) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie vorzugsweise Ethandiol, Butandiol, Hexandiol oder besonders bevorzugt Butandiol und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, bevorzugt mit 5 oder 6 C-Atomen im cycloaliphatischen Ring, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, bevorzugt C_3-C_{12}-Alkyldiolen, wie beispielsweise Neopentyglykol, und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen wie beispielsweise 1,2,3-Propantriol oder Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren, vorzugsweise C_2-C_{12}-Alkyldicarbonsäuren, wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder

F) aus säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,

oder einer Mischung und/oder einem Copolymer aus E und F, wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;

aliphatische oder teilaromatische Polyesterurethane aus

G) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie beispielsweise Ethandiol, Butandiol, Hexandiol, besonders bevorzugt Butandiol und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, vorzugsweise mit C_5- oder C_6-cycloaliphatischem Ring, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkyldiolen, wie beispielsweise Neopentyglykol, und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkylpolyolen, wie beispielsweise 1,2,3-Propantriol oder Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren, vorzugsweise C_2-C_{12}-Alkyldicarbonsäuren, wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure, und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder

H) aus säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen, beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,

oder einer Mischung und/oder einem Copolymer aus G und H, wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;

I) aliphatischen und/oder cycloaliphatischen bifunktionellen und zusätzlich gegebenenfalls höherfunktionellen Isocyanaten, mit vorzugsweise 1 bis 12 C-Atomen bzw. 5 bis 8 C-Atomen im Falle von cycloaliphatischen Isocyanaten, z.B. Tetramethylendiisocyanat, Hexamethylendiisocyanat, Isophorondiisocyanat, gegebenenfalls zusätzlich mit linearen und/oder verzweigten und/oder cycloaliphatischen bifunktionellen und/oder höherfunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkyldi- oder -polyole bzw. 5 bis 8 C-Atomen im Falle von cycloaliphatischen Alkoholen, z.B. Ethandiol, Hexandiol, Butandiol, Cyclohexandimethanol, und / oder gegebenenfalls zusätzlich mit linearen und/oder verzweigten und/oder cycloaliphatischen bifunktionellen und/oder höherfunktionellen Aminen und / oder Aminoalkoholen mit vorzugsweise 2 bis 12 C-Atomen in der Alkylkette, z.B. Ethylendiamin oder Aminoethanol, und / oder gegebenenfalls weitere modifizierte Amine oder Alkohole wie beispielsweise Ethylendiaminoethansulfonsäure, als freie Säure oder als Salz,

wobei der aus G) und H) gebildete Esteranteil mindestens 75 Gew.-%, bezogen auf die Summe aus G), H) und J), beträgt;

aliphatische oder aliphatisch-aromatische Polyestercarbonate aus

J) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie beispielsweise Ethandiol, Butandiol, Hexandiol oder besonders bevorzugt Butandiol und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, vorzugsweise mit 5 bis 8 C-Atomen im cycloaliphatischen Ring, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Mole-

kulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, vorzugsweise mit C_2-C_{12}-Alkyldicarbonsäuren, wie beispielsweise Neopentyglykol und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen wie beispielsweise 1,2,3-Propantriol, Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder

K) säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette, beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,

oder einer Mischung und/oder einem Copolymer aus J) und K), wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;

und

L) einem Carbonatanteil, der aus aromatischen bifunktionellen Phenolen, bevorzugt Bisphenol-A, und Carbonatspendern, beispielsweise Phosgen, hergestellt wird

oder

einem Carbonatanteil, der aus aliphatischen Kohlensäureestern oder deren Derivaten wie beispielsweise Chlorkohlensäureestern oder aliphatischen Carbonsäuren oder deren Derivaten wie beispielsweise Salzen und Carbonatspendern, beispielsweise Phosgen, hergestellt wird, wobei

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der aus J) und/oder K) gebildete Esteranteil mindestens 70 Gew.-%, bezogen auf die Summe aus J), K) und L) beträgt;

aliphatische oder teilaromatische Polyesteramide aus

A) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie beispielsweise Ethandiol, Butandiol, Hexandiol, besonders bevorzugt Butandiol, und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, vorzugsweise mit 5 bis 8 C-Atomen, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkyldiolen, wie beispielsweise Neopentyglykol und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkylpolyole, wie beispielsweise 1,2,3-Propantriol, Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette, wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder

B) aus säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen in der Kohlenstoffkette, beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,

oder einer Mischung und/oder einem Copolymer aus A) und B), wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;

und

C) aliphatischen und/oder cycloaliphatischen bifunktionellen und/oder gegebenenfalls geringen Mengen verzweigten bifunktionellen Aminen, bevorzugt sind lineare aliphatische C_2 bis C_{10}-Diamine, und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Aminen, unter den Aminen bevorzugt Hexamethylendiamin, Isophorondiamin und besonders bevorzugt Hexamethylendiamin, sowie aus linearen und/oder cycloaliphatischen bifunktionellen Säuren, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette bzw. C_5- oder C_6-Ring im Falle von cycloaliphatischen Säuren, bevorzugt Adipinsäure, und/oder ggf. geringen Mengen von verzweigten bifunktionellen und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren, vorzugsweise mit 2 bis 10 C-Atomen, oder

D) säure- und aminfunktionalisierten Bausteinen, vorzugsweise mit 4 bis 20 C-Atomen in der cycloaliphatischen Kette, bevorzugt ω-Laurinlactam, ε-Caprolactam, besonders bevorzugt ε-Caprolactam,

oder einer Mischung aus C) und D), wobei der aus A) und/oder B) gebildete Esteranteil mindestens 20 Gew.-%, bezogen auf die Summe aus A), B), C) und D) beträgt, vorzugsweise der aus A) und/oder B) gebildete Esteranteil 20 bis 85 Gew.-%, und der Anteil der aus C) und/oder D) gebildeten Amidstrukturen 80 bis 15 Gew.-% beträgt.

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Bei den in der erfindungsgemäßen Folie verwendeten biologisch abbaubaren Polymeren kann es sich sowohl um reine Polymere als auch um Mischungen aus verschiedenen Polymeren handeln, wobei im Falle von Mischungen vorzugsweise Polymere aus nur einer der vorgenannten Verbindungsklassen (Polyester, Polyesterurethane, Polyestercarbonate, Polyesteramide) verwendet werden.

Die für die erfindungsgemäße ein- oder mehrschichtige Folie eingesetzten Polymere können zusätzlich übliche Additive und Hilfsmittel enthalten.

Bevorzugt kommen in den für die erfindungsgemäße Folie eingesetzten Polymeren folgende Additive und Hilfsmittel zum Einsatz:

Maximal 5 Gew.-%, z.B. 0 bis 5 Gew.-% für Polyester typisch eingesetzte Nukleierungsmittel (beispielsweise 1,5-Naphthalindinatriumsulfonat oder Schichtsilikate, beispielsweise Talkum, oder Keimbildner der Nanoteilchengröße, d. h. mittlerer Teilchendurchmesser < 1 µm, aus beispielsweise Titannitrid, Aluminiumhydroxylhydrat, Bariumsulfat oder Zirkonverbindungen) und/oder maximal 5 Gew.-%, z.B. 0 bis 5 Gew.-% der üblichen Stabilisatoren und Neutralisationsmittel und/oder maximal 5 Gew.-%, z.B. 0 bis 5 Gew.-% der üblichen Gleit- und Trennmittel und/oder maximal 5 Gew.-%, z.B. 0 bis 5 Gew.-% der üblichen Antiblockmittel sowie gegebenenfalls Pigmente zur Einfärbung.

Als Stabilisatoren und Neutralisationsmittel können die üblichen stabilisierend wirkenden Verbindungen für Polyesterverbindungen eingesetzt werden. Deren Zusatzmenge liegt 0 maximal bei 5 Gew.-%.

Besonders geeignet als Stabilisatoren sind phenolische Stabilisatoren, Alkali-/Erdalkalistearate und/oder Alkali-/Erdalkalicarbonate. Phenolische Stabilisatoren werden in einer Menge von 0 bis 3 Gew.-%, insbesondere 0,15 bis 0,3 Gew.-% und mit einer Molmasse von mehr als 500 g/mol bevorzugt. Pentaerythrityl-Tetrakis-

3(3,5-di-Tertiärbutyl-4-Hydroxyphenyl)-Propionat oder 1,3,5-Trimethyl-2,4,6-tris-(3,5-di-Tertiärbutyl-4-Hydroxybenzyl)-benzol sind besonders vorteilhaft.

Neutralisationsmittel sind vorzugsweise Dihydrotalcit, Calciumstearat, Calciumcarbonat und/oder Calciummontanat einer mittleren Teilchengröße von höchstens 0,7 µm, einer absoluten Teilchengröße von kleiner 10 µm und einer spezifischen Oberfläche von mindestens 40 m^2/g.

In einer besonders bevorzugten Aufführungsform der Folie besitzt diese einen Nukleierungsmittelanteil von 0,0001 bis 2 Gew.-% und einen Stabilisatoren- und Neutralisationsmittelanteil von 0,0001 bis 2 Gew.-%.

Gleit- und Trennmittel sind höhere aliphatische Amide, tertiäre Amine, aliphatische Säureamide, höhere aliphatische Säureester, niedrigmolekulare polarmodifizierte Wachse, Montanwachse, cyclische Wachse, Phthalate, Metallseifen sowie Silikonöle. Besonders geeignet ist der Zusatz von höheren aliphatischen Säureamiden und Silikonölen.

Unter den aliphatischen Amiden sind insbesondere die Angebotsformen von Ethylenamid bis Stearylamid geeeignet. Aliphatische Säureamide sind Amide einer wasserunlöslichen Monocarbonsäure (sogenannte Fettsäuren) mit 8 bis 24 Kohlenstoffatomen, vorzugsweise 10 bis 18 Kohlenstoffatomen. Erucasäureamid, Stearinsäureamid und Ölsäureamid sind hierunter bevorzugt.

Geeignet als Trenn- oder Gleitmittel sind außerdem Verbindungen, die sowohl Ester- als auch Amidgruppen enthalten, wie beispielsweise Stearamid-Ethylstearat bzw. 2 Stear-Amido-Ethyl-Stearat.

Unter der Bezeichnung Montanwachse fällt ein Reihe von verschiedenen Verbindungen. Siehe hierzu Neumüller et al. in Römpps Chemie-Lexikon, Franckh'sche Verlagshandlung, Stuttgart, 1974.

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Als cyclische Wachse sind beispielsweise Komponenten wie cyclische Adipinsäuretetramethylenester bzw. 1.6-Dioxa-2.7-dioxocyclododecan, oder das homologe Hexamethylenderivat geeignet. Solche Stoffe sind als Handelsprodukte mit dem Namen Glycolube VL bekannt.

Geeignete Silikonöle sind Polydialkylsiloxane, vorzugsweise Polydimethylsiloxan, Polymethylphenylsiloxan, olefinmodifiziertes Silikon, mit Polyethern modifiziertes Silikon wie z. B. Polyethylenglykol und Polypropylenglykol sowie epoxyamino- und alkoholmodifiziertes Silikon. Die Viskosität der geeigneten Silikonöle liegt im Bereich von 5 000 bis 1 000 000 mm^2/s. Polydimenthylsiloxan mit einer Viskosität von 10.000 bis 100.000 mm^2/s ist bevorzugt.

Die Menge des zugesetzten Gleitmittels beträgt maximal 5 Gew.-%. In einer besonders bevorzugten Ausführungsform der Folie besitzt diese einen Gleitmittelanteil von 0,005 bis 4 Gew.-%. In einer ganz besonders bevorzugten Ausführungsform der Folie besitzt diese einen Gleitmittelanteil von 0,05 bis 1 Gew.-%.

Geeignete Antiblockmittel sind sowohl anorganische als auch organische Zusatzstoffe, die aufgrund ihrer Teilchengröße und/oder Form als Erhebung aus der Folienoberfläche herausragen und somit einen Abstandshaltereffekt hervorrufen.

In einer bevorzugten Form werden als anorganische Antiblockmittel die folgenden Stoffe eingesetzt:

Aluminiumhydroxid
Aluminiumsilikate, beispielsweise Kaolin oder Kaolinton,
Aluminiumoxide, beispielsweise Θ-Aluminiumoxid
Aluminiumsulfat
Keramiken aus Silica-Aluminiumoxiden
Bariumsulfat

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natürliche und synthetische Kieselsäuren
Schichtsilikate, beispielsweise Asbest,
Siliciumdioxid
Calciumcarbonat vom Calcit-Typ
Calciumphosphat
Magnesiumsilikate
Magnesiumcarbonat
Magnesiumoxid
Titandioxid
Zinkoxid
Microglaskugeln
und als organische Antiblockmittel die folgenden Stoffe eingesetzt:
mit dem biologisch abbaubaren Polymer unverträgliche organische Polymerisate wie Stärke
Polystyrole
Polyamide
Polycarbonate
vernetztes und unvernetztes Polymethylmethacrylat
vernetztes Polysiloxan (z.B. Tospearl)
polarmodifizierts Polyethylen (z.B. Maleinsäureanhydrid-gepfropftes Polyethylen)
polarmodifizierts Polypropylen (z.B. Maleinsäureanhydrid-gepfropftes Polypropylen)
statistische Copolymer auf Ethylen- oder Propylenbasis mit Vinylalcohol oder Vinylacetat oder Acrylsäure oder Acrylsäureester oder Methacrylsäure oder Methacrylsäureester oder Metallsalzen der Methacrylsäure oder Metallsalze der Methacrylsäureester
Benzoguanamin Formaldehyd Polymere
aliphatische und teilaromatische Polyester mit anderen Schmelzpunkten als der Folienrohstoff
aliphatische Polyesteramide mit anderen Schmelzpunkten als der Folienrohstoff
aliphatische Polyesterurethane mit anderen Schmelzpunkten als der Folienrohstoff

aliphatisch-aromatische Polyestercarbonate mit anderen Schmelzpunkten als der Folienrohstoff

Die wirksame Menge an Antiblockmittel liegt im Bereich von 0 bis maximal 5 Gew.-%. In einer besonders bevorzugten Ausführung enthält die Folie 0,005 bis 4 Gew.-% Antiblockmittel. In einer ganz besonders bevorzugten Ausführung enthält die Folie 0,05 bis 1 Gew.-% Antiblockmittel. Die mittlere Teilchengröße liegt zwischen 1 und 6 µm, insbesondere 2 und 5 µm, wobei Teilchen mit einer kugelförmigen Gestalt, wie in der EP-A-0 236 945 und der DE-A-38 01 535 beschrieben, besonders geeignet sind. Besonders geeignet sind auch Kombinationen verschiedener Abstandshaltersysteme. In einer besonders bevorzugten Ausführung werden die Abstandshaltersysteme den äußeren Deckschichten zugegeben.

Die erfindungsgemäße biologisch abbaubare und kompostierbare ein- oder mehrschichtige Folie mit verbesserten Barriereeigenschaften kann Pigmente zur Einfärbung enthalten. Dabei kann es sich um organische oder anorganische Pigmente, die der Deckschicht, der Zwischenschicht oder den Zwischenschichten oder allen Schichten zugegeben werden. In einer besonders bevorzugten Form sind die Pigmente biologisch unbedenklich und/oder haben eine Konzentration von weniger als 1 Gew.-%.

Zur Herstellung der erfindungsgemäßen Folie werden die Polymere für die gegebenenfalls mit Additiven versehene Folie bzw. die einzelnen Folienschichten gemäß dem üblichen Stand der Technik mit den gewünschten Gewichtsmengen an Adsorbens sowie organischen oder anorganischen Additiven und/oder Hilfsstoffen ausgerüstet. Dies geschieht beim Granulieren des Rohstoffs beispielsweise in Zweischneckenextrudern, wo dem Rohstoff die Additive und/oder Absorbentien zugegeben werden. Neben dieser Art der Additivierung und Zugabe der Absorbentien besteht auch die Möglichkeit, daß ein Teil oder alle notwendigen Additive und/oder Absorbentien einem nicht oder teilweise ausgerüsteten Rohstoff in Form eines Masterbatches zugegeben werden. Unter dem Begriff Masterbatch ist im Rahmen der

vorliegenden Erfindung eine Stammischung zu verstehen, insbesondere ein granulatförmiges staubfreies Konzentrat eines Kunststoffrohstoffes mit hohen Mengen an Additiven und/oder Absorbentien, das in der Masseaufbereitung als Zwischenprodukt verwendet wird (als Materialzusatz zu einem nicht oder nur teilweise oder unvollständig mit Additiven und/oder Absorbentien ausgerüstetem Granulat), um daraus Folien herzustellen, die eine bestimmte Menge an Additiven enthalten. Das Masterbatch wird vor dem Einfüllen des Polymergranulates in den Extruder in derartigen Mengen zu den nicht oder nur teilweise oder unvollständig mit Additiven und/oder Absorbentien ausgerüsteten Rohstoffen zugemischt, so daß die gewünschten Gewichtsprozentanteile an Additiven und/oder Absorbentien in den Folien realisiert werden. Die Materialien, aus denen neben den Additiven und/oder Absorbentien die Masterbatche hergestellt werden, sind in einer besonders bevorzugten Form ebenfalls biologisch abbaubar, biologisch unbedenklich, oder haben einen Gewichtsanteil, der kleiner als 1 Gew.-% ist.

Die bevorzugten Materialien, aus denen neben den Additiven die Masterbatche hergestellt werden, sind Stoffe, die mit den in dieser Erfindung genannten Polymeren verträglich sind.

Die Herstellung der erfindungsgemäßen ein- oder mehrschichtigen Folie geschieht zweckmäßig über ein Extrusionsverfahren. Die in Granulatform, gegebenenfalls einschließlich der gewünschten Additive und Hilfsstoffe, vorliegenden Polymere werden in einem oder mehreren Extrudern aufgeschmolzen, homogenisiert, komprimiert und über eine Ein- oder Mehrschichtdüse ausgetragen. Dabei kann es sich bei der Düse um eine Ringdüse zur Herstellung einer nahtlosen Schlauchfolie oder eine Flachdüse zur Herstellung einer Flachfolie handeln. Die so ausgetragene bzw. z.B. mittels Walzenpresseuren ausgezogene Folie wird anschließend bis zur Verfestigung abgekühlt. Die Kühlung kann dabei sowohl über Luft als auch über Wasser oder auch mittels Kühlwalzen erfolgen. Die Kühlung kann dabei einseitig oder beidseitig, im Falle einer Schlauchfolie innen- und außenseitig oder nur innen- oder nue außenseitig

erfolgen. Die Schlauchfolie kann zudem einseitig oder beidseitig geschnitten werden, so daß man eine mehrschichtige Flachfolie erhält.

Die Folie kann weiterhin nach der Abkühlung bei teilkristallinen Werkstoffen unter Kristallitschmelztemperatur und bei amorphen Werkstoffen oberhalb der Glasübergangstemperatur temperiert und dann ein oder mehrfach monoaxial oder biaxial verstreckt werden, wobei das biaxiale Verstrecken nacheinander oder, insbesondere im Fall von Schlauchfolien, simultan erfolgen kann. Nach der oder den Verstreckstufen kann jeweils optional eine Fixierng der Folie durch eine Wärmebehandlung erfolgen. Ein besonders geeignetes Verstreckverfahren ist dabei für das simultane biaxiale Recken von Schlauchfolien die sogenannte Double-Bubble-Technologie, bei der die Verstreckung einer Primärblase über einen anliegenden Innendruck erfolgt. Zur gezielten Einstellung von Schrumpfeigenschaften kann die Folie anschließend einer Wärmebehandlung unterzogen werden. Die Folie kann in diesem Fixierprozess wieder bis kurz unter die Kristallitschmelztemperatur erwärmt werden.

Sofern anschließend eine Coronabehandlung erfolgen soll, wird zwecksmäßigerweise so vorgegangen, daß die Folie zwischen zwei als Elektroden dienenden Leiterelementen hindurchgeführt wird, wobei zwischen den Elektroden eine so hohe Spannung, meist Wechselspannung (etwa 5 bis 20 kV und 5 bis 30 kHz), angelegt ist, daß Sprüh- oder Coronaentladungen stattfinden können. Durch die Sprüh- oder Coronaentladung wird die Luft oberhalb der Folienoberfläche ionisiert und reagiert mit den Molekülen der Folienoberfläche, so daß zusätzlich polare Einlagerungen in der Polymermatrix entstehen.

Für eine gegebenenfalls an den Reck- bzw. Fixierprozeß anschließende Flammbehandlung mit polarisierter Flamme (vgl. US-A-4,622,237) wird eine elektrische Gleichspannung zwischen einem Brenner (negativer Pol) und einer Kühlwalze angelegt. Die Höhe der angelegten Spannung beträgt zwischen 400 und 3 000 V, vorzugsweise liegt sie im Bereich von 500 bis 2 000 V. Durch die angelegte Spannung erhalten die ionisierten Atome eine erhöhte Beschleunigung und treffen mit größerer

kinetischer Energie auf die Polymeroberfläche. Die chemischen Bindungen innerhalb des Polymermoleküls werden leichter aufgebrochen und die Radikalbildung geht schneller vonstatten. Die thermische Belastung des Polymeren ist hierbei weitaus geringer als bei der Standardflammbehandlung, und es können Folien erhalten werden, bei denen die Siegeleigenschaften der behandelten Seite sogar besser sind als diejenigen der nicht behandelten Seite.

Bei einer sich gegebenenfalls an den Reck- bzw. Fixierprozeß anschließende Plasmavorbehandlung werde in einer Niederdruckkammer Gase, z. B. Sauerstoff oder Stickstoff oder Kohlendioxid oder Methan oder Halogenkohlenwasserstoffe oder Silanverbindungen oder höhermolekulare Verbindungen oder auch Mischungen hiervon, einem hochenergiereichen Feld, z. B. Mikrowellenstrahlung, ausgesetzt. Es entstehen hochenergiereiche Elektronen, die auf die Moleküle treffen und ihre Energien übertragen. Hierdurch entstehen lokal radikalische Strukturen, deren Anregungszustände Temperaturen von einigen zehntausend Grad Celsius entsprechen, obwohl das Plasma selbst sich nahezu auf Raumtemperatur befindet. Dadurch besteht die Möglichkeit, chemische Bindungen aufzubrechen und Reaktionen in Gang zu setzen, die normalerweise erst bei hohen Temperaturen ablaufen können. Es bilden sich Monomerradikale und Ionen. Aus den entstandenen Monomerradikalen bilden sich - teilweise schon im Plasma - kurzkettige Oligomere, die dann auf der zu beschichtenden Oberfläche kondensieren und polymerisieren. Auf dem Beschichtungsgut scheidet sich dadurch ein homogener Film ab.

Bevor sich jedoch die Monomerradikale bzw. Ionen auf dem Substrat abscheiden, besteht außerdem zusätzlich die Möglichkeit, in der sogenannten After-Glow-Zone einen weiteren Stoffstrom den angeregten Molekülen hinzuzufügen. Hierdurch ist es möglich, einen Stoff oder ein Stoffgemisch zu erzeugen, das bei dem Auftreffen auf der Polymerfilmoberfläche einen oxidativen Angriff bei den Substratpolymeren erzeugt. Es bildet sich eine glasartige und meist hochvernetzte Schicht, die mit der Folienoberfläche fest verbunden ist. Bei geeigneter Stoffzusammensetzung ergibt sich hierdurch einen Anstieg der Oberflächenspannung auf der Folie.

Zudem können bei mehrschichtigen Ausführungsformen der erfindungsgemäßen Folie weitere Adsorbens enthaltene oder Adsorbens-freie Folienschichten durch einen Kaschierprozeß, bei dem die Kaschierfolie als Zulauf durch Naß- oder Thermokaschierung auf die ursprüngliche Folie aufgebracht wird, hergestellt werden.

Bei der Naßkaschierung wird die ursprüngliche Folie oder die Kaschierfolie oder sowohl ursprüngliche Folie als auch Kaschierfolie mit einem Kaschierkleber benetzt, beide Folien durch Walzenpresseure miteinander verbunden und anschließend getrocknet. Als Kaschierkleber kommen sowohl lösungsmittelfreie als auch lösungsmittelhaltige Kleber in Betracht. In einer besonders bevorzugten Form ist der Kaschierkleber ebenfalls biologisch abbaubar und kompostierbar.

Bei der Thermokaschierung wird als zulaufende Kaschierfolie eine Folie verwendet, die durch temperierte Walzenpresseure auf die beschichtete Folie gepreßt wird und damit mit der beschichtete Folienseite verbunden wird. In einer besonders bevorzugten Form wird als zulaufende Kaschierfolie eine mehrschichtige Folie verwendet, die aus mindestens zwei Schichten bestehen, deren Schmelzpunkte verschieden sind. Durch Temperieren der Kaschierfolie auf die Schmelz- oder Erweichungstemperatur der früher schmelzenden Schicht und Anpressen auf die ursprüngliche Folienseite wird die Verbindung zwischen beschichteter Folie und Kaschierfolie erreicht, wobei die früher schmelzende Schicht der Kaschierfolie der ursprünglichen Seite zugewandt ist.

Die Kaschierfolie, die durch Naß- oder Thermokaschierung mit der ursprünglichen Folie verbunden wird, kann sowohl ungereckt, monoaxial als auch biaxial gereckt sein.

Gegenstand der Erfindung ist darüber hinaus die Anwendung der erfindungsgemäßen Folie. Als bevorzugte Anwendung kommt der Einsatz dieser Folie insbesondere als mehrschichtige Folie in vorbehandelter oder unvorbehandelter sowie in bedruckter,

unbedruckter oder lackierter Form für die Verpackung insbesondere von geruchsintensiven Lebensmitteln oder Nichtlebensmitteln oder für Schutz- und Trennfunktionen bei Kosmetik und Hygieneartikeln, beispielsweise für Babywindeln oder Damenbinden in Betracht.

Die folgenden Beispiele sollen die Erfindung weiter erläutern, ohne sie darauf zu beschränken:

Beispiele

Beispiel 1 (Vergleichsbeispiel)

Aus zwei unterschiedlichen biologisch abbaubaren und kompostierbaren Polymeren wurde an einer Drei-Schicht-Blasfolienanlage ein symmetrischer, coextrudierter Film mit einem ABA-Aufbau hergestellt. Das Material A Polyesteramid aus 58 Gew.-% Caprolactam, 23 Gew.-% Adipinsäure, 19 % Butandiol, (LP BAK 403-004, Bayer AG) hat einen MFI von 9 (in g/10min bei 190°C, 2,16 kg, gemessen nach DIN 53 735), einen Schmelzpunkt von 125°C, gemessen nach ISO 3146 / C2, einen Anteil an Gleitmittel von 0,15 Gew.-% und einen Antiblockanteil bzw. Nukleierungsmittelgehalt von 0,1 Gew.-%. Das Material B Polyesteramid aus 62 Gew.-% Caprolactam, 21 Gew.-% Adipinsäure, 17 Gew.-% Butandiol (LP BAK 404-002, Bayer AG) hat einen MFI von 6 (in g/10min bei 190°C, 2,16 kg, gemessen nach DIN 53 735), einen Schmelzpunkt von 140°C, gemessen nach ISO 3146 / C2 und einen Antiblockanteil bzw. Nukleierungsmittelgehalt von 0,1 Gew.-%. Die maximale Extrusionstemperatur betrug 190°C. Die Schmelze wurde über eine Mehrschichtringdüse ausgetragen und durch Luft abgekühlt. Die maximale Düsentemperatur betrug 185°C. Es wurde in diesem Fall eine Folie mit einer Gesamtdicke von 80 µm in einer Breite von 700mm hergestellt, wobei das Material A mit dem niedrigen Schmelzpunkt die Außenschichten bildet.

Beispiel 2

Aus den gleichen biologisch abbaubaren Polymeren A und B aus Beispiel 1 wurde an der gleichen Blasfolienanlage unter Berücksichtigung der selben Prozeßparametern ein symetrischer ABA-Film hergestellt. Im Gegensatz zum Beispiel 1 wurde dem Material B der innenliegenden Kernschicht 20 Gew.-% eines Masterbatches zugegeben, das 18,4 Gew.-% des Absorbers Mordenit Natrium-Form aktiv (Hersteller: Bayer AG) enthält. Es wurde ein Film mit einer Dicke von 75 µm hergestellt.

Beispiel 3

Aus den biologisch abbaubaren Polymeren A und B aus Beispiel 1 wurde an der gleichen Blasfolienanlage unter Berücksichtigung der selben Prozeßparametern ein symetrischer ABA-Film hergestellt. Im Gegensatz zum Beispiel 1 und 2 wurde dem Material B der innenliegenden Kernschicht 20 Gew.-% eines Masterbatches zugegeben, das 18,8 Gew.-% des Absorbers Zeolith-X Natrium-Form aktiv (Hersteller: Bayer AG) enthält. Es wurde ein Film mit einer Dicke von 80 µm hergestellt.

Beispiel 4

Aus den biologisch abbaubaren Polymeren A und B aus Beispiel 1 wurde an der gleichen Blasfolienanlage unter Berücksichtigung der selben Prozeßparametern ein symetrischer ABA-Film hergestellt. Im Gegensatz zum Beispiel 1, 2 und 3 wurde dem Material B der innenliegenden Kernschicht 20 Gew.-% eines Masterbatches zugegeben, das 19,4 Gew.-% des Absorbers Zeolith-A Calcium-, Natrium-Form aktiv (Hersteller: Bayer AG) enthält. Es wurde ein Film mit einer Dicke von 80 µm hergestellt.

An den gefertigten Mustern wurden die folgenden physikalischen Eigenschaften wie folgt gemessen:

Mechanische Eigenschaften:

An den Proben wurden die mechanischen Größen Reißfestigkeit und Reißdehnung sowohl in Längs- als auch in Querrichtung gemäß DIN 53 455 bestimmt. Der E-Modul in Längs- und Querrichtung wurde entsprechend der DIN 53 457 gemessen. Die Dicke der einzelnen Muster wurde nach DIN 53 370 bestimmt.

Permeationseigenschaften:

An den Proben wurde die Durchlässigkeit von Sauerstoff bei einer Temperatur von 23°C und einer relativen Feuchte von 0% und 75% nach DIN 53 380 gemessen. Weiterhin wurde die Durchlässigkeit von Wasserdampf bei einer Temperatur von 23°C und einer relativen Feuchte von 85% gemäß DIN 53 122 bestimmt.

Sensorik-Test

Ein Becherglas wird mit frischen kleingehackten Zwiebel gefüllt, Folien nach Beispiel 1 bis 4 werden so über ein Becherglas gelegt, daß sie die Öffnung vollständig abschließen.

Mittels sensorischer Geruchsprobe wird der Unterschied in der Gasdurchlässigkeit bestimmt.

Folie nach Beispiel 1: Sofort ist der Geruch der Zwiebeln durch die Folie zu riechen.

Folien nach Beispiel 2 - 4: Der Geruch der feingehackten Zwiebeln ist nicht wahrzunehmen, d.h. der Absorber nimmt die Geruchsbestandteile auf und läßt sie nicht durch die Folie.

Siegelnahtfestigkeiten:

An den Proben wurde die Festigkeit von Siegelnähten, die an einem Siegelgerät mit beidseitig beheiztem Siegelwerkzeug gefertigt wurden, bestimmt. Unter der Siegelfestigkeit wird die Höchstkraft verstanden, die erforderlich ist, um eine unter definierten Bedingungen (Druck, Zeit, Temperatur) hergestellte Siegelnaht zu trennen. Die Siegelnaht wird in N angegeben, die Streifenbreite als Index hinzu-

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gefügt (N/15mm). Der Siegelbereich ist der Temperaturbereich, in dem eine meßbare Siegelfestigkeit vorhanden ist.

Aus der zu prüfenden Folienbahn wurden zwei fehlerfreie, saubere Probenstreifen entnommen. Zur Siegelung wurden sie mit den zu versiegelnden Flächen aufeinandergelegt und so zwischen die Siegelbacken gehalten, daß die Probe an jeder Seite mindestens 1cm überstand. Die Siegelung muß senkrecht zur Folienlaufrichtung erfolgen.

Die Siegelfestigkeitsprüfung erfolgte nach folgender Standardbedingung:

Druck:	50N/cm²
Zeit:	0,5sec
Temperatur:	in 10K-Schritten ab 150°C.

Aus der hergestellten Siegelnaht wurde aus der Mitte exakt in Folienlaufrichtung mit einem Prüfstreifenschneider ein 15mm breiter Prüfstreifen geschnitten. Mit einer Zugfestigkeitsmaschine wurde er auf Siegelfestigkeit untersucht, indem er senkrecht zur Siegelnaht getrennt wurde. Angegeben wurde jeweils der Maximalwert der beim Zerreißen auftretenden Kraft.

Die Ergebnisse der Untersuchungen an den Proben aus Beispiel 1, 2, 3 und 4 sind in der Tabelle aufgeführt.

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	Beispiel 1	Beispiel 2	Beispiel 3	Beispiel 4
Mechanische Eigenschaften				
Dicke [µm]	80	75	80	80
E-Modul längs [MPa]	213	218	232	227
E-Modul quer [Mpa]	303	290	356	330
Reißfestigkeit längs [MPa]	35	34	33	31
Reißfestigkeit quer [MPa]	24	22	18	20
Reißdehnung längs [%]	406	437	363	368
Reißdehnung quer [%]	639	563	522	545
Permeation				
Sauerstoff 23 °C/0 % r. F. [cm³/m²/d/bar]	247	231	234	204
Sauerstoff 23 °C/75 % r. F. [cm³/m²/d/bar]	341	365	355	306
Wasserdampf 23 °C/85 % r. F. [g/m2/d]	120	128	145	130
Sensorik-Test				
wahrnehmbarer Zwiebelgeruch	ja	nein	nein	nein
Siegelfestigkeit				
Notwendige Zerreißkraft [N/15mm]	29 (bei 160°C)	31 (bei 160°C)	25 (bei 160°C)	25 (bei 160°C)
Biologisch abbaubar				
gemäß DIN V 54900	ja	ja	ja	ja

Patentansprüche

1. Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige Folie aus einem oder mehreren biologisch abbaubaren und kompostierbaren Polymeren und üblichen Additiven und/oder Hilfsstoffen, dadurch gekennzeichnet, daß die Folie in mindestens einer Schicht mindestens ein Absorbens zur Bindung gasförmiger Geruchs- oder Aromastoffe enthält.

2. Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige Folie gemäß Anspruch 1, dadurch gekennzeichnet, daß als Absorbens geruchsabsorbierende Feststoffe, insbesondere Zeolithe, eingesetzt werden.

3. Biologisch abbaubare und kompostierbare, ein oder mehrschichtige Folie gemäß Anspruch 1 oder 2, dadurch gekennzeichnet, daß der Gehalt an Absorbens in der mindestens ein Absorbens enthaltenden Folienschicht oder den mindestens ein Absorbens enthaltenen Folienschichten 0,01 bis 25 Gew.-%, insbesondere 0,01 bis 10 Gew.-%, bezogen auf das Gesamtgewicht der jeweiligen Folienschicht, beträgt.

4. Biologisch abbaubare und kompostierbare Folie gemäß einem der Anspüchen 1 bis 3, dadurch gekennzeichnet, daß die Folie mindestens dreischichtig ist und die beiden äußeren Folienschichten kein Absorbens enthalten.

5. Biologisch abbaubare und kompostierbare Folie gemäß einem der Ansprüche 1 bis 4, dadurch gekennzeichnet, daß die Folie mehrschichtig ist und in mindestens zwei Schichten unterschiedliche Absorbentien enthält.

6. Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige Folie gemäß einem der Ansprüche 1 bis 5, dadurch gekennzeichnet, daß die Folie ungereckt oder monoaxial gereckt oder biaxial gereckt ist.

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7. Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige Folie gemäß einem der Ansprüche 1 bis 6, dadurch gekennzeichnet, dass es die biologisch abbaubaren und kompostierbaren Polymere, Polyesterpolymere aus der Gruppe Polyester, Polyesterurethane, Polyestercarbonate, Polyesteramide sind.

8. Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige Folie gemäß einem der Ansprüche 1 bis 7, dadurch gekennzeichnet, daß es sich bei den biologisch abbaubaren und kompostierbaren Polymeren um

aliphatische oder teilaromatische Polyesteramide aus

A) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie beispielsweise Ethandiol, Butandiol, Hexandiol, besonders bevorzugt Butandiol, und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, vorzugsweise mit 5 bis 8 C-Atomen, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkyldiolen, wie beispielsweise Neopentyglykol und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkylpolyole, wie beispielsweise 1,2,3-Propantriol, Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette, wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenen-

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falls geringen Mengen höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder

B) aus säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen in der Kohlenstoffkette, beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,

oder einer Mischung und/oder einem Copolymer aus A) und B), wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;

und

C) aliphatischen und/oder cycloaliphatischen bifunktionellen und/oder gegebenenfalls geringen Mengen verzweigten bifunktionellen Aminen, bevorzugt sind lineare aliphatische C_2 bis C_{10}-Diamine, und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Aminen, unter den Aminen bevorzugt Hexamethylendiamin, Isophorondiamin und besonders bevorzugt Hexamethylendiamin, sowie aus linearen und/oder cycloaliphatischen bifunktionellen Säuren, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette bzw. C_5- oder C_6-Ring im Falle von cycloaliphatischen Säuren, bevorzugt Adipinsäure, und/oder ggf. geringen Mengen von verzweigten bifunktionellen und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren, vorzugsweise mit 2 bis 10 C-Atomen, oder

D) säure- und aminfunktionalisierten Bausteinen, vorzugsweise mit 4 bis 20 C-Atomen in der cycloaliphatischen Kette, bevorzugt ω-Laurinlactam, ε-Caprolactam, besonders bevorzugt ε-Caprolactam,

oder einer Mischung aus C) und D), wobei der aus A) und/oder B) gebildete Esteranteil mindestens 20 Gew.-%, bezogen auf die Summe aus A), B), C) und D) beträgt, vorzugsweise der aus A) und/oder B) gebildete Esteranteil 20 bis 85 Gew.-% und der Anteil der aus C) und/oder D) gebildeten Amidstrukturen 80 bis 15 Gew.-% beträgt, oder

aliphatische oder teilaromatische Polyester aus

E) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie beispielsweise Ethandiol, Butandiol, Hexandiol oder besonders bevorzugt Butandiol und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, bevorzugt mit 5 oder 6 C-Atomen im cycloaliphatischen Ring, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, bevorzugt C_3-C_{12}-Alkyldiolen, wie beispielsweise Neopentyglykol, und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen wie beispielsweise 1,2,3-Propantriol oder Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren, vorzugsweise C_2-C_{12}-Alkyldicarbonsäuren, wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphtha-

lindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder

F) aus säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,

oder einer Mischung und/oder einem Copolymer aus E) und F), wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;

aliphatische oder teilaromatische Polyesterurethane aus

G) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie beispielsweise Ethandiol, Butandiol, Hexandiol, besonders bevorzugt Butandiol und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, vorzugsweise mit C_5- oder C_6-cycloaliphatischem Ring, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, vorzugsweise C_3-C_{12}Alkyldiolen, wie beispielsweise Neopentyglykol, und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkylpolyolen, wie beispielsweise 1,2,3-Propantriol oder Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren, vorzugsweise C_2-C_{12}-Alkyldicarbonsäuren, wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure, und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthal-

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säure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder

H) aus säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen, beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,

oder einer Mischung und/oder einem Copolymer aus G) und H), wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;

und

I) aliphatischen und/oder cycloaliphatischen bifunktionellen und zusätzlich gegebenenfalls höherfunktionellen Isocyanaten, mit vorzugsweise 1 bis 12 C-Atomen bzw. 5 bis 8 C-Atomen im Falle von cycloaliphatischen Isocyanaten, z.B. Tetramethylendiisocyanat, Hexamethylendiisocyanat, Isophorondiisocyanat, gegebenenfalls zusätzlich mit linearen und/oder verzweigten und/oder cycloaliphatischen bifunktionellen und/oder höherfunktionellen Alkoholen, vorzugsweise C_3-C_{12}-Alkyldi- oder -polyole bzw. 5 bis 8 C-Atomen im Falle von cycloaliphatischen Alkoholen, z.B. Ethandiol, Hexandiol, Butandiol, Cyclohexandimethanol, und/oder gegebenenfalls zusätzlich mit linearen und/oder verzweigten und/oder cycloaliphatischen bifunktionellen und/oder höherfunktionellen Aminen und/oder Aminoalkoholen mit vorzugsweise 2 bis 12 C-Atomen in der Alkylkette, z.B. Ethylendiamin oder Aminoethanol, und/oder gegebenenfalls weitere modifizierte Amine oder Alkohole wie beispielsweise Ethylendiaminoethansulfonsäure, als freie Säure oder als Salz,

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wobei der aus G) und/oder H) gebildete Esteranteil mindestens 75 Gew.-%, bezogen auf die Summe aus G), H) und D), beträgt;

aliphatische oder aliphatisch-aromatische Polyestercarbonate aus

J) aliphatischen bifunktionellen Alkoholen, bevorzugt linearen C_2 bis C_{10}-Dialkoholen wie beispielsweise Ethandiol, Butandiol, Hexandiol oder besonders bevorzugt Butandiol und/oder gegebenenfalls cycloaliphatischen bifunktionellen Alkoholen, vorzugsweise mit 5 bis 8 C-Atomen im cycloaliphatischen Ring, wie beispielsweise Cyclohexandimethanol, und/oder teilweise oder vollständig statt der Diole monomere oder oligomere Polyole auf Basis Ethylenglykol, Propylenglykol, Tetrahydrofuran oder Copolymere daraus mit Molekulargewichten bis 4000, bevorzugt bis 1000, und/oder gegebenenfalls geringen Mengen von verzweigten bifunktionellen Alkoholen, vorzugsweise mit C_2-C_{12}-Alkyldicarbonsäuren, wie beispielsweise Neopentyglykol und zusätzlich gegebenenfalls geringen Mengen von höherfunktionellen Alkoholen wie beispielsweise 1,2,3-Propantriol, Trimethylolpropan sowie aus aliphatischen bifunktionellen Säuren wie beispielsweise und bevorzugt Bernsteinsäure, Adipinsäure und/oder gegebenenfalls aromatischen bifunktionellen Säuren wie beispielsweise Terephthalsäure, Isophthalsäure, Naphthalindicarbonsäure und zusätzlich gegebenenfalls geringen Mengen höherfunktionellen Säuren wie beispielsweise Trimellitsäure oder

K) säure- und alkoholfunktionalisierten Bausteinen, vorzugsweise mit 2 bis 12 C-Atomen in der Alkylkette, beispielsweise Hydroxybuttersäure, Hydroxyvaleriansäure, Milchsäure, oder deren Derivaten, beispielsweise ε-Caprolacton oder Dilactid,

oder einer Mischung und/oder einem Copolymer aus J) und K), wobei die aromatischen Säuren nicht mehr als 50 Gew.-% Anteil, bezogen auf alle Säuren, ausmachen;

und

L) einem Carbonatanteil, der aus aromatischen bifunktionellen Phenolen, bevorzugt Bisphenol-A, und Carbonatspendern, beispielsweise Phosgen, hergestellt wird
oder
einem Carbonatanteil, der aus aliphatischen Kohlensäureestern oder deren Derivaten wie beispielsweise Chlorkohlensäureestern oder aliphatischen Carbonsäuren oder deren Derivaten wie beispielsweise Salzen und Carbonatspendern, beispielsweise Phosgen, hergestellt wird, wobei

der aus J) und /oder K) gebildete Esteranteil mindestens 70 Gew.-%, bezogen auf die Summe aus J), K) und L) beträgt;

handelt.

9. Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige Folie gemäß einem der Ansprüche 1 bis 8, dadurch gekennzeichnet, daß es sich bei den erfindungsgemäßen biologisch abbaubaren und kompostierbaren Polymeren sowohl um reine Polymere als auch um Mischungen aus verschiedenen Polymeren handeln kann.

10. Biologisch abbaubare und kompostierbare, ein- oder mehrschichtige gemäß einem der Ansprüche 1 bis 9, dadurch gekennzeichnet, daß als Additive und/oder Hilfsmittel 0 bis maximal 5 Gew.-% Nukleierungsmitteln und 0 bis maximal 5 Gew.-% der üblichen Stabilisatoren und Neutralisationsmittel und

0 bis maximal 5 Gew.-% der üblichen Gleit- und Trennmittel und Pigmente zur Einfärbung der Folie in einer, mehreren oder allen Schichten sowie 0 bis maximal 5 Gew.-% der üblichen Antiblockmittel bevorzugt in einer oder den äußeren Deckschichten enthält, wobei sich die Konzentrationsangaben auf die Gesamtmasse der Folie beziehen.

11. Verwendung der biologisch abbaubaren und kompostierbaren, ein- oder mehrschichtigen Folie gemäß einem der Ansprüche 1 bis 10 in vorbehandelter oder unvorbehandelter sowie in bedruckter, unbedruckter oder lackierter Form für die Verpackung von Lebensmitteln oder Wasserdampf- oder Sauerstoff-empfindlichen oder geruchsintensiven Nichtlebensmitteln.

12. Verwendung der biologisch abbaubaren und kompostierbaren, ein- oder mehrschichtigen Folie gemäß einem der Ansprüche 1 bis 10 in Kosmetik oder Hygieneartikeln.

INTERNATIONAL SEARCH REPORT

Inter nal Application No
PCT/EP 00/10769

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C08J5/18 B32B27/08 C08K3/22

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 7 B32B C08J C08K

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 practical, search terms used)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	DE 198 11 773 A (WOLFF WALSRODE AG) 23 September 1999 (1999-09-23) claims	1-12
Y	EP 0 811 392 A (PROCTER & GAMBLE) 10 December 1997 (1997-12-10) claims 1,5	1-12

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in 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 document 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
24 January 2001	06/02/2001

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	De Jonge, S

Form PCT/ISA/210 (second sheet) (July 1992)

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
PCT/EP 00/10769

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
DE 19811773 A	23-09-1999	AU 3328699 A	11-10-1999
		WO 9947602 A	23-09-1999
EP 0811392 A	10-12-1997	EP 0811390 A	10-12-1997
		EP 0811391 A	10-12-1997
		AU 3147197 A	05-01-1998
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		CA 2257628 A	11-12-1997
		CN 1225569 A	11-08-1999
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INTERNATIONALER RECHERCHENBERICHT

Internales Aktenzeichen
PCT/EP 00/10769

A. KLASSIFIZIERUNG DES ANMELDUNGSGEGENSTANDES
IPK 7 C08J5/18 B32B27/08 C08K3/22

Nach der Internationalen Patentklassifikation (IPK) oder nach der nationalen Klassifikation und der IPK

B. RECHERCHIERTE GEBIETE

Recherchierter Mindestprüfstoff (Klassifikationssystem und Klassifikationssymbole)
IPK 7 B32B C08J C08K

Recherchierte aber nicht zum Mindestprüfstoff gehörende Veröffentlichungen, soweit diese unter die recherchierten Gebiete fallen

Während der internationalen Recherche konsultierte elektronische Datenbank (Name der Datenbank und evtl. verwendete Suchbegriffe)
EPO-Internal, WPI Data

C. ALS WESENTLICH ANGESEHENE UNTERLAGEN

Kategorie*	Bezeichnung der Veröffentlichung, soweit erforderlich unter Angabe der in Betracht kommenden Teile	Betr. Anspruch Nr.
Y	DE 198 11 773 A (WOLFF WALSRODE AG) 23. September 1999 (1999-09-23) Ansprüche	1-12
Y	EP 0 811 392 A (PROCTER & GAMBLE) 10. Dezember 1997 (1997-12-10) Ansprüche 1,5	1-12

☐ Weitere Veröffentlichungen sind der Fortsetzung von Feld C zu entnehmen

☒ Siehe Anhang Patentfamilie

* Besondere Kategorien von angegebenen Veröffentlichungen :
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"X" Veröffentlichung von besonderer Bedeutung; die beanspruchte Erfindung kann allein aufgrund dieser Veröffentlichung nicht als neu oder auf erfinderischer Tätigkeit beruhend betrachtet werden
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"&" Veröffentlichung, die Mitglied derselben Patentfamilie ist

Datum des Abschlusses der internationalen Recherche	Absendedatum des internationalen Recherchenberichts
24. Januar 2001	06/02/2001

Name und Postanschrift der Internationalen Recherchenbehörde	Bevollmächtigter Bediensteter
Europäisches Patentamt, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	De Jonge, S

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INTERNATIONALER RECHERCHENBERICHT

Angaben zu Veröffentlichungen, die zur selben Patentfamilie gehören

Interr ales Aktenzeichen
PCT/EP 00/10769

Im Recherchenbericht angeführtes Patentdokument	Datum der Veröffentlichung	Mitglied(er) der Patentfamilie	Datum der Veröffentlichung
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		WO 9947602 A	23-09-1999
EP 0811392 A	10-12-1997	EP 0811390 A	10-12-1997
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		EP 0912149 A	06-05-1999
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Ｂ２９Ｃ　45/14		7344－4Ｆ		
51/00		7722－4Ｆ		
Ｂ６５Ｄ　1/09				
		6671－3Ｅ	Ｂ６５Ｄ　1／00　Ｂ	

審査請求　未請求　請求項の数１(全　4　頁)　最終頁に続く

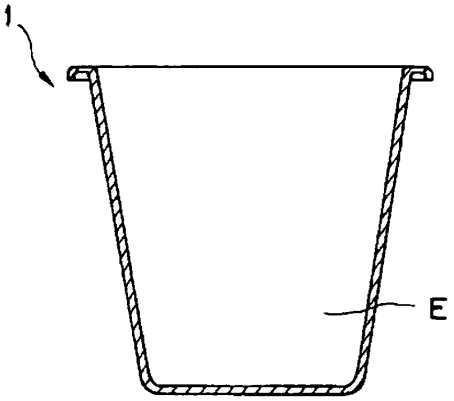
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(54)【発明の名称】　食品用容器

(57)【要約】
【目的】　射出成形法または熱成形法で形成された分解性を備える容器を提供する。
【構成】　射出成形法または熱成形法で形成された容器は、分解性材料の単層または積層体であるように構成する。



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【特許請求の範囲】

【請求項1】　射出成形法または熱成形法で形成された食品を収納する容器であって、該容器は分解性材料の単層または積層体から形成されていることを特徴とする食品用容器。

【発明の詳細な説明】

【0001】

【産業上の利用分野】本発明は、食品を収納する容器、特に、射出成形法または熱成形法で形成された容器であって、かつ分解性を有する材料からなる食品容器に関する。

【0002】

【従来の技術】近年、プラスッチック容器はガラス瓶、缶などに比べ、軽くて丈夫で輸送・保管に便利でコストが安価なためその需要は益々拡大している。このようなプラスッチック容器の中には、収納する内容物として食品のみを対象とし、射出成形法または熱成形法で形成された容器がある。このような容器は一般の日常生活に密着して頻繁に使用されており、その需要は年々拡大している。

【0003】ところで、このような射出成形法または熱成形法で形成された容器を含めたプラスチック容器の使用後の処理に関しては従来、焼却ないし埋め立て等に頼らざるを得ず、プラスチック廃棄物処理は、現在大きな社会問題としてクローズアップされて来ている。すなわち、焼却処理では、包装材料廃棄物の大きな燃焼エネルギーに耐え得る耐高熱炉が必要になり、処理コストが高いものとなる。また、埋め立て処理では、プラスチック製の包装材料は分解せずそのままの形態で地中に存在するので、埋め立て地の地盤が安定しないという問題がある。さらに、地上に散乱した包装材料は、分解性がないため半永久的にゴミとして残り、環境を損なうという問題がある。

【0004】このような問題を解決するために、いわゆる分解性を有するプラスチック材料の使用の提案がなされている。

【0005】

【発明が解決しようとする課題】しかしながら、従来の提案のものでは、収納する内容物として食品のみを対象とし、射出成形法または熱成形法で形成された容器であり、かつ、分解性を有する食品容器は存在していなかった。このように実情のもの本発明は創案されたものであって、その目的は、射出成形法または熱成形法で形成され、かつ分解性を備える食品容器を提供することにある。

【0006】

【課題を解決するための手段】上記課題を解決するために、本発明は、射出成形法または熱成形法で形成された食品を収納する容器であって、該容器は分解性材料の単層または積層体から形成されているように構成した。

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【0007】

【作用】本発明の食品容器は分解性材料からなるので、通常の放置処理がなされても自然に分解する。

【0008】

【実施例】以下、本発明の実施例を図1乃至図3に基づいて具体的に説明する。本発明の食品容器は、食品を収納する容器であって、特に、射出成形法または熱成形法で形成された容器が対象になる。射出成形容器は、粒状の熱可塑性の樹脂を加熱シリンダの一端に送り込み、プランジャの前進によりシリンダ内に圧縮、加熱して流動性となし、ノズルから冷えた金型の中に射出圧入して、冷却固化させて成形する方法である。

【0009】熱成形法は、熱可塑性プラスチックシートを加熱軟化させ、真空や圧空を利用して金型の形状に形成する方法である。このように形成された食品容器は、カップ形状のものやトレー状のもの等があり、図1にはカップ形状の食品容器1が示される。このような容器1は食品を収納させるための器状の収納部Eを有し、容器そのものは、分解性材料の単層または積層体から形成される。

【0010】単層形態の場合、分解性材料としては、分解性プラスチック、特に、分解性ポリエチレン樹脂や分解性ポリプロピレン樹脂等の分解性ポリオレフィン樹脂、ポリビニルアルコールまたは分解性ポリエステルで形成される。分解性ポリオレフィン脂は、ポリエチレンやポリプロピレンを主成分とし光や微生物により分解され得る樹脂である。

【0011】分解性ポリオレフィン樹脂のうち、光分解性のものとしては、エチレンと一酸化炭素との共重合体等が挙げられる。このエチレン・一酸化炭素共重合体は、カルボニル基の結合する2番目と3番目の炭素間が光で開裂することにより分解すると言われている。そして、分解速度は共重合体中の一酸化炭素の含有量により調節することができる。通常、エチレン・一酸化炭素共重合体の密度は0．８９～0．９５g／cm³程度であり、一酸化炭素の含有量は0．１～１０モル％程度である。

【0012】上述のようなエチレン・一酸化炭素共重合体は、例えばエチレンと一酸化炭素とを温度２３０℃、圧力２０００気圧程度の条件下で共存させることにより製造することができる。また、光分解性の分解性ポリオレフィン樹脂として、ポリエチレン（密度0．８７０～0．９５０g／cm³、溶融指数（ＭＦＩ）0．４～４０）、ポリプロピレン（密度0．８８～0．９１g／cm³、溶融指数（ＭＦＩ）0．２～６０）と有機酸金属塩との混合物を用いることもできる。金属化合物としては、ステアリン酸鉄、ステアリン酸セリウム、ステアリン酸コバルト等があり、酸化鉄等の金属酸化物、アセチルアセトンやジブチルジチオカーバメートなどとの錯体等が挙げられ、これら金属化合物の混合量は１～５００

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0ppm 程度が好ましい。

【0013】また、ビニルケトンとの共重合体を添加することもある。また、分解性ポリオレフィン樹脂のうち、微生物分解性のものとしては、ポリエチレンやポリプロピレンと、ポリカプロラクトン、デンプンおよび微生物により重合されるポリエステル等との混合物が挙げられる。生分解性ポリオレフィン樹脂に用いられるポリエチレンとしては、密度0．870～0．950g／cm³、溶融指数（MFI）0．4～40のエチレンの単独重合体、もしくはプロピレン、ヘキセン、オクテン、4－メチルペンテン－1等の他のオレフィンとのランダムあるいはブロック共重合体、さらには酢酸ビニル、アクリル酸、メタクリル酸、アクリル酸メチル、アクリル酸エチル、メタクリル酸メチル、無水マレイン酸等のエチレン性不飽和基を有する単量体との共重合体等が挙げられる。ポリプロピレンとしては密度0．88～0．91g／cm³、溶融指数（MFI）0．2～60のプロピレンとエチレン、ブテン－1、無水マレイン酸等のエチレン性不飽和基を有する単量体との共重合体等が挙げられる。

【0014】また、生分解性ポリエチレン樹脂に用いられるポリカプロラクトンは、ε－カプロラクトンの開環重合により得られ、その重量平均分子量（Mw）は、通常、40，000～100，000程度である。また、デンプンは、D－グルコースの重合体であり、ジャガイモ、サツマイモ、トウモロコシ、小麦等の茎や根から工業的に製造されるものであり、その重量平均分子量（Mw）は原料、製法により数万から数千万まで大きく変化する。このようなデンプンの平均粒径は10μm以下が好ましい。

【0015】生分解性ポリエチレン樹脂に用いられる微生物により重合されるポリエステルとしては、3－ヒドロキシブチレートと3－ヒドロキシバリレートとのランダム共重合ポリエステル（例えば、英国I.C.I.社により生産されている水素細菌にプロピオン酸を供給して得られるもの）や、水素細菌に吉草酸を供給して得られる3－ヒドロキシブチレート主体のポリエステル等が挙げられる。

【0016】上述のような生分解性ポリエチレン樹脂におけるポリカプロラクトン、デンプンおよび微生物により重合されるポリエステルとの混合量は、ポリオレフィンと上記各混合成分の合計を100重量%として、それぞれ5～80重量%が好ましい。上記各成分の混合量が5重量%未満では、微生物分解性が不十分であり、また80重量%を越えると強度的に弱くなってしまう。

【0017】なお、ポリカプロラクトン、デンプンおよび微生物により重合されるポリエステルの各成分は2種以上を用いてもよいが、その場合、混合量は合計で5～80重量%の範囲となればよい。また、微生物分解性のポリオレフィン樹脂の原料として前述の光分解性のポリ

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オレフィン樹脂を用いることにより、光分解性と微生物分解性を兼ね備えた分解性ポリエチレン樹脂を得ることができる。

【0018】分解性ポリエステルとしては、前述のポリカプロラクトン、微生物により重合されるポリエステルを単独で用いることができる。また、ポリビニルアルコールとしては、ケン化度99%以上のものが通常使用される。これらにも前記の無機物含有させてもよい。なお、このような分解性のポリエチレン樹脂の中には、強度を上げるために、炭酸カルシウム、炭酸マグネシウム、硫酸カルシウム、亜硫酸カルシウム、硫酸バリウム、硫酸マグネシウム等の金属塩、ケイ酸あるいはカオリン、タルク等のケイ酸塩、酸化チタン、酸化亜鉛等の金属酸化物および水酸化アルミニウム、アルミナ等のアルミニウム化合物等の無機質充填剤を含有させてもよい。さらに酸化防止剤、分解促進剤、安定剤、帯電防止剤、界面活性剤等の各種添加剤を含有させてもよい。

【0019】積層体の場合の断面の種々の態様が図2または図3に示される。いずれも分解性を有する材質からなっている。図2には、積層体の一実施例としての部分断面図が示される。図2に示されるように容器1は、2層の積層構造からなり、例えば、内層21を上記の分解性ポリオレフィン樹脂、外層25をポリビニルアルコールまたは分解性ポリエステルで形成させる。これらの2層の間には熱融着性のよい他の樹脂を接着層として介在させてもよい。内層21の厚さは、通常、10～2000μm程度である。また、外層25の厚さは、通常、10～2000μm程度である。

【0020】図3には、積層体の他の一実施例としての部分断面図が示される。図3に示されるように容器1は、3層の積層構造からなり、内層21および外層35が中間層30を挟持するように形成されている。この場合、内層21および外層35をそれぞれ上記分解性ポリオレフィン樹脂から形成し、中間層30をポリビニルアルコールまたは分解性ポリエステルで形成させる。これらの層間には熱融着性のよい他の樹脂を接着層として介在させてもよい。内層21および外層35の厚さは、通常、10～2000μm程度である。また、中間層30の厚さは、通常、10～2000μm程度である。

【0021】その他、上記分解性プラスチックの種々のものを、適宜、組み合わせて種々の積層体を形成することもできる。

【0022】

【発明の効果】本発明の容器は分解性材料からなるので、分解性を備えるという効果を奏する。

【図面の簡単な説明】

【図1】本発明の容器の斜視図である。

【図2】本発明の容器の層構成を示す部分断面図である。

【図3】本発明の容器の他の層構成を示す部分断面図で

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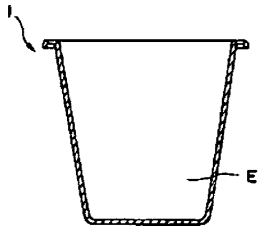
5　　6

ある。

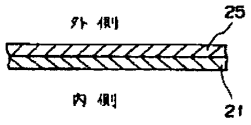
1　容器

【符号の説明】

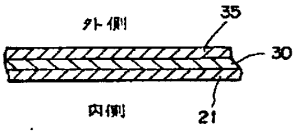
【図1】



【図2】



【図3】



フロントページの続き

(51)Int.Cl.⁵	識別記号	庁内整理番号	FI	技術表示箇所
// B32B 27/00		H 7717－4F		
B29K 23:00				
29:00				
B29L 31:00		4F		

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PATENT ABSTRACTS OF JAPAN

(11)Publication number : 04-299131
(43)Date of publication of application : 22.10.1992

(51)Int.Cl. B29D 31/00
B29C 45/14
B29C 51/00
B65D 1/09
// B32B 27/00
B29K 23:00
B29K 29:00
B29L 31:00

(21)Application number : 03-064900
(22)Date of filing : 28.03.1991

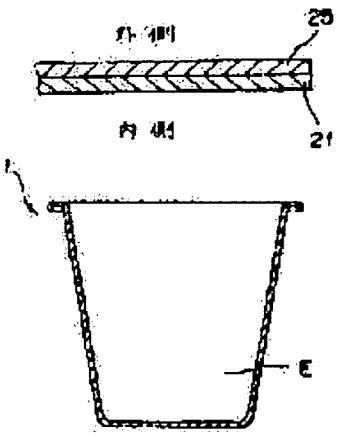
(71)Applicant : DAINIPPON PRINTING CO LTD
(72)Inventor : SUGIYAMA YASUHARU
TSUCHIYA HIROTAKA

(54) VESSEL FOR FOOD

(57)Abstract:

PURPOSE: To enable natural decomposition of vessels scattered about on the ground, by a method wherein the vessels holding food which are molded by an injection molding or thermoforming method are formed of a single layer or laminated body of a decomposable material decomposed by light or microbes.

CONSTITUTION: In a cuppy or traylike vessel 1 for food which is formed by an injection molding method or a thermoforming method, a single layer or a laminated layer of a decomposable material is used as its material. In case of a form of the single, layer, the decomposable material is regarded as a polyolefin resin such as decomposable polyethylene resin and decomposable polypropylene or polyvinyl alcohol or decomposable polyester. In case of the laminated body, for example, an inner layer 21 is regarded as decomposable polyolefin resin and an outer layer 25 is regarded as the polyvinyl alcohol or the decomposable polyester.



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LEGAL STATUS

[Date of request for examination]

[Date of sending the examiner's decision of rejection]

[Kind of final disposal of application other than the examiner's decision of rejection or application converted registration]

[Date of final disposal for application]

[Patent number]

[Date of registration]

[Number of appeal against examiner's decision of rejection]

[Date of requesting appeal against examiner's decision of rejection]

[Date of extinction of right]

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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
11/348,150	02/06/2006	Christopher S. Cleveland	FDS-029388-US	5681

1726 7590 07/16/2008

INTERNATIONAL PAPER COMPANY
6285 TRI-RIDGE BOULEVARD
LOVELAND, OH 45140

EXAMINER	
PATTERSON, MARC A	
ART UNIT	PAPER NUMBER
1794	

MAIL DATE	DELIVERY MODE
07/16/2008	PAPER

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.

Office Action Summary	Application No.	Applicant(s)	
	11/348,150	CLEVELAND ET AL.	
	Examiner	Art Unit	
	MARC A. PATTERSON	1794	

-- 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 1 MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, 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 _____.

2a)☐ This action is **FINAL**. 2b)☒ This action is non-final.

3)☐ 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

4)☒ Claim(s) 1-73 is/are pending in the application.

4a) Of the above claim(s) _____ is/are withdrawn from consideration.

5)☐ Claim(s) _____ is/are allowed.

6)☐ Claim(s) _____ is/are rejected.

7)☐ Claim(s) _____ is/are objected to.

8)☒ Claim(s) 1-73 are subject to restriction and/or election requirement.

Application Papers

9)☐ The specification is objected to by the Examiner.

10)☐ 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).

11)☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

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).

a)☐ All b)☐ Some * c)☐ None of:

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) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
3) ☐ Information Disclosure Statement(s) (PTO/SB/08) Paper No(s)/Mail Date _____.
4) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date. _____ .
5) ☐ Notice of Informal Patent Application
6) ☐ Other: _____.

DETAILED ACTION

Election/Restrictions

1. Restriction to one of the following inventions is required under 35 U.S.C. 121:

I. Claims 1 – 58, drawn to a laminate with oxygen barrier properties, classified in class 428, subclass 35.7.

II. Claims 59 – 73, drawn to a method for making biodegradable paper products, classified in class 264, subclass 177.2.

2. Inventions I and II are related as process of making and product made. The inventions are distinct if either or both of the following can be shown: (1) that the process as claimed can be used to make another and materially different product or (2) that the product as claimed can be made by another and materially different process (MPEP § 806.05(f)). In the instant case the product can be made by another and materially different process, such as a method in which a barrier layer is laminated instead of extruded.Restriction for examination purposes as indicated is proper because all these inventions listed in this action are independent or distinct for the reasons given above and there would be a serious search and examination burden if restriction were not required because one or more of the following reasons apply:

(a) the inventions have acquired a separate status in the art in view of their different classification;

(b) the inventions have acquired a separate status in the art due to their recognized divergent subject matter;

(c) the inventions require a different field of search (for example, searching different classes/subclasses or electronic resources, or employing different search queries);

(d) the prior art applicable to one invention would not likely be applicable to another invention;

(e) the inventions are likely to raise different non-prior art issues under 35 U.S.C. 101 and/or 35 U.S.C. 112, first paragraph.

Applicant is advised that the reply to this requirement to be complete must include (i) an election of a invention to be examined even though the requirement may be traversed (37 CFR 1.143) **and (ii) identification of the claims encompassing the elected invention**.

The election of an invention may be made with or without traverse. To reserve a right to petition, the election must be made with traverse. If the reply does not distinctly and specifically point out supposed errors in the restriction requirement, the election shall be treated as an election without traverse. Traversal must be presented at the time of election in order to be considered timely. Failure to timely traverse the requirement will result in the loss of right to petition under 37 CFR 1.144. If claims are added after the election, applicant must indicate which of these claims are readable on the elected invention.

If claims are added after the election, applicant must indicate which of these claims are readable upon the elected invention.

Should applicant traverse on the ground that the inventions are not patentably distinct, applicant should submit evidence or identify such evidence now of record showing the inventions to be obvious variants or clearly admit on the record that this is the case. In either

instance, if the examiner finds one of the inventions unpatentable over the prior art, the evidence or admission may be used in a rejection under 35 U.S.C. 103(a) of the other invention.

3. This application contains claims directed to the following patentably distinct species.

i. A laminate comprising a polymeric protectant layer, moisture protective layer and degradable tie layer (Claims 1 – 35, 38 and 41 – 43).

ii. A laminate with oxygen barrier properties comprising a second degradable tie layer (Claim 36).

iii. A laminate with oxygen barrier properties comprising a fourth polyester layer (Claim 37).

iv. A laminate with oxygen barrier properties comprising three degradable tie layers (Claim 39).

v. A laminate with oxygen barrier properties comprising a third degradable tie layer disposed on a moisture protectant layer (Claim 40).

vi. A laminate for forming moisture barriers comprising two moisture protectant layers (Claims 44 – 58)

The species are independent or distinct because claims to the different species recite the mutually exclusive characteristics of such species. In addition, these species are not obvious variants of each other based on the current record.

Applicant is required under 35 U.S.C. 121 to elect a single disclosed species for prosecution on the merits to which the claims shall be restricted if no generic claim is finally held to be allowable. Currently, Claims 1, 36 – 37, 39 – 40 and 44 are generic.

There is an examination and search burden for these patentably distinct species due to their mutually exclusive characteristics. The species require a different field of search (e.g., searching different classes/subclasses or electronic resources, or employing different search queries); and/or the prior art applicable to one species would not likely be applicable to another species; and/or the species are likely to raise different non-prior art issues under 35 U.S.C. 101 and/or 35 U.S.C. 112, first paragraph.

Applicant is advised that the reply to this requirement to be complete must include (i) an election of a species to be examined even though the requirement may be traversed (37 CFR 1.143) **and (ii) identification of the claims encompassing the elected species**, including any claims subsequently added. An argument that a claim is allowable or that all claims are generic is considered nonresponsive unless accompanied by an election.

The election of the species may be made with or without traverse. To preserve a right to petition, the election must be made with traverse. If the reply does not distinctly and specifically point out supposed errors in the election of species requirement, the election shall be treated as an election without traverse. Traversal must be presented at the time of election in order to be considered timely. Failure to timely traverse the requirement will result in the loss of right to petition under 37 CFR 1.144. If claims are added after the election, applicant must indicate which of these claims are readable on the elected species.

Should applicant traverse on the ground that the species are not patentably distinct, applicant should submit evidence or identify such evidence now of record showing the species to be obvious variants or clearly admit on the record that this is the case. In either instance, if the

examiner finds one of the species unpatentable over the prior art, the evidence or admission may be used in a rejection under 35 U.S.C. 103(a) of the other species.

Upon the allowance of a generic claim, applicant will be entitled to consideration of claims to additional species which depend from or otherwise require all the limitations of an allowable generic claim as provided by 37 CFR 1.141.

4. A telephone call was made to Mr. Thomas W. Barnes on April 23, 2008 to request an oral election to the above restriction requirement, but did not result in an election being made.

5. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Marc A Patterson whose telephone number is 571-272-1497. The examiner can normally be reached on Mon - Fri 8:30 AM - 5:00 PM.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Rena Dye can be reached on 571-272-1498. 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 http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free).

/Marc A Patterson/
Primary Examiner, Art Unit 1794

Application Number	Application/Control No.	Applicant(s)/Patent under Reexamination	
	11/348,150	CLEVELAND ET AL.	
	Examiner	Art Unit	
	MARC A. PATTERSON	1794	

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INTERNATIONAL PAPER

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F 513 248 6455

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CONFIDENTIAL FACSIMILE TRANSMISSION

To:	COMMISSIONER FOR PATENTS
Fax:	571-273-8300
Tel:	
Date:	AUGUST 18, 2008
RE:	RESTRICTION REQUIREMENTS

Our File Ref.: FDS-029388-US
Your File Ref.: 11/348,150
No. of Pages: 5 (including this cover sheet)

☐ URGENT ☐ FOR YOUR INFORMATION

☐ CONFIRMATION COPY FOLLOWS BY:
- ☐ FIRST-CLASS U.S. MAIL
- ☐ OVERNIGHT MAIL
- ☐ INTERNATIONAL MAIL

☒ NO COPY FOLLOWS

In response to the Office Action dated 7/16/2008, please find the following checked items:

☐ Petition for Extension of Time Under 37 C.F.R. 1.136(a), Form PTO/SB/22, and one (1) copy thereof attached thereto, _____ sheet(s);

☒ Response to Restriction Requirement, 3 sheet(s);

☐ Fee Transmittal, Form PTO/SB/17, _____ sheet(s); and,

☐ Information Disclosure Statement by Applicant(s), _____ sheet(s), to which Form PTO/SB/08, _____ sheet(s), has been attached along with a copy of each reference(s) cited thereon.

Thank you.

Certificate of Transmission Under 37 C.F.R. 1.8

The undersigned hereby certifies that a true and accurate copy of the items checked above are being transmitted to the Honorable Commissioner for Patents, by facsimile transmission to the facsimile number indicated above, on this the 18th day of August, 20 08

JAMES A. TOMLINSON

This facsimile transmission may contain confidential and/or legally privileged information from the law department of International Paper Company which is intended only for the use of the individual(s) named above. If you are not the intended recipient, you are hereby notified that any disclosure, copying or distribution of this facsimile transmission is strictly prohibited. If you have received this transmission in error, please notify us immediately by phoning (513) 248-6207 so that we can arrange for the return of the documents. Thank you.

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MATTHEW M. ESLAMI, PE
PATENT AGENT
INTELLECTUAL PROPERTY

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LOVELAND OH 45140

T 513 248 6193
F 513 248 6455
matthew.eslami@ipaper.com

SENT BY ☒ FACSIMILE TO (571-273-8300)
☐ FIRST CLASS MAIL TO THE ADDRESS BELOW

August 18, 2008

Commissioner for Patents
Post Office Box 1450
Alexandria, Virginia 22313-1450

RE: RESPONSE TO RESTRICTION REQUIREMENT
Applicant : CLEVELAND, et al
Serial No. : 11/348,150
Filed on : FEBRUARY 6, 2006
Title : BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOISTURE.....
Our Ref. : FDS-029388-US

Dear Commissioner:

Enclosed herewith for filing, Applicant(s) respectfully submit(s) the following checked items:

☐ Petition for Extension of Time Under 37 C.F.R. 1.136(a), Form PTO/SB/22, and one (1) copy thereof attached thereto, _____ sheet(s);

☒ Response to Restriction Requirement, __3__ sheet(s);

☐ Fee Transmittal, Form PTO/SB/17, _____ sheet(s);

☐ Information Disclosure Statement by Applicant(s), _____ sheet(s), to which Form PTO/SB/08, _____ sheet(s), has been attached along with a copy of each reference(s) cited thereon; and,

☐ a postage-prepaid, return-receipt postcard with a listing of the above-checked items.

Please stamp the enclosed postcard and return same to me to indicate your receipt of the above-listed items. Please feel free to contact me if you have any questions concerning the above or the enclosed.

With kindest regards,

Matthew M. Eslami
Agent for Applicant(s)

Enclosures
MME/jat

CERTIFICATE OF MAILING OR TRANSMISSION UNDER 37 C.F.R. § 1.8

The undersigned hereby certifies that a true and accurate copy of the documents checked hereon, together with all attachments referred to herein, is being transmitted to the Honorable Commissioner for Patents, either by first-class mail, postage prepaid, addressed to Commissioner for Patents, Post Office Box 1450, Alexandria, Virginia 22313-1450, or by facsimile transmission to the facsimile number indicated hereon, on this the 18th day of AUGUST 20 08

JANE A. TOMLINSON

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Docket No.: FDS-029388-US

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF

CLEVELAND, et al. : GROUP ART UNIT: 1794

SERIAL NO: 11/348,150 : EXAMINER: PATTERSON, MARC A

FILED: February 06, 2006 :

FOR: BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOISTURE BARRIER PROPERTIES AND METHOD FOR MAKING BIODEGRADABLE PAPER-BASED LAMINATE

RESPONSE TO RESTRICTION REQUIREMENT

COMMISSIONER FOR PATENTS
UNITED STATES PATENT AND TRADEMARK OFFICE
PO BOX 1450
ALEXANDRIA VA 22313-1450

SIR:

Responsive to the Official Communication dated July 16, 2008, Applicants elect, with traverse, Group I, Claims 1-58; drawn to a laminate with oxygen barrier properties, classified in class 428, subclass 35.7 for further prosecution.

PAGE 3/5 * RCVD AT 8/18/2008 11:43:54 AM [Eastern Daylight Time] * SVR:USPTO-EFXRF-4/0 * DNIS:2738300 * CSID:513 248 6451 * DURATION (mm-ss):02-00

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REMARKS

The Office has required restriction in the present application as follows:

Group I: Claims 1-58; drawn to a laminate with oxygen barrier properties, classified in class 428, subclass 35.7.

Group II: Claims 59-73, drawn to a method of making biodegradable paper products, classified in class 264, subclass 177.2.

Applicants elect, with traverse, Group I, Claims 1-58; drawn to a laminate with oxygen barrier properties, for further prosecution.

The Office has characterized the inventions of Groups I and II as related as process of making and product made. Citing MPEP §806.05(f), the Office concludes that the process as claimed can be used to make other and materially different product. However, there is no evidence of record to show that the claimed product for making biodegradable paper products with barrier properties can be made from materially different process such as extrusion as the Office has alleged. Applicants respectfully submit that the Office has not shown that "making biodegradable lids for trays" are materially different from the claimed product. The Office has not provided sufficient reasons and/or examples to support this assertion. The Office has merely stated the conclusion. Accordingly, the Office has failed to meet the burden necessary in order to sustain the Restriction Requirement. Accordingly, Applicants respectfully submit that the Restriction Requirement should be withdrawn.

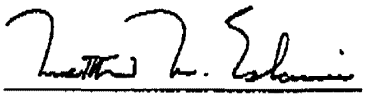
PAGE 4/5 * RCVD AT 8/18/2008 11:43:54 AM [Eastern Daylight Time] * SVR:USPTO-EFXRF-4/0 * DNIS:2738300 * CSID:513 248 6451 * DURATION (mm-ss):02-00

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Finally, Applicants respectfully remind the Office that if the Office finds the traversed restriction requirement addressed herein as final and that the claimed product as elected herein as patentable, the Office is compelled to rejoin the claims drawn to the method of making such claimed product. Early and favorable indication is respectfully requested.

The Applicants further submit that this application is in condition for examination on the merits and an early notification to that effect is earnestly solicited.

Date: August 18, 2008

By: 

Matthew M. Eslami
Reg. No. 45,488
Patent Agent for Applicant
International Paper Company
6285 Tri-Ridge Boulevard
Loveland, Ohio 45140
Tel: (513) 248-6193
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e-mail: matt.eslami@ipaper.com

PAGE 5/5 * RCVD AT 8/18/2008 11:43:54 AM [Eastern Daylight Time] * SVR:USPTO-EFXRF-4/0 * DNIS:2738300 * CSID:513 248 6451 * DURATION (mm-ss):02-00 AT P.005

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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
11/348,150	02/06/2006	Christopher S. Cleveland	FDS-029388-US	5681

1726 7590 12/02/2008

INTERNATIONAL PAPER COMPANY
6285 TRI-RIDGE BOULEVARD
LOVELAND, OH 45140

EXAMINER	
PATTERSON, MARC A	
ART UNIT	PAPER NUMBER
1794	

MAIL DATE	DELIVERY MODE
12/02/2008	PAPER

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.

Office Action Summary	Application No.	Applicant(s)	
	11/348,150	CLEVELAND ET AL.	
	Examiner	Art Unit	
	MARC A. PATTERSON	1794	

-- 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 1 MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, 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 _____.

2a)☐ This action is **FINAL**. 2b)☒ This action is non-final.

3)☐ 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

4)☒ Claim(s) 1-73 is/are pending in the application.

4a) Of the above claim(s) _____ is/are withdrawn from consideration.

5)☐ Claim(s) _____ is/are allowed.

6)☐ Claim(s) _____ is/are rejected.

7)☐ Claim(s) _____ is/are objected to.

8)☒ Claim(s) 1-73 are subject to restriction and/or election requirement.

Application Papers

9)☐ The specification is objected to by the Examiner.

10)☐ 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).

11)☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

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).

a)☐ All b)☐ Some * c)☐ None of:

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) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
3) ☐ Information Disclosure Statement(s) (PTO/SB/08) Paper No(s)/Mail Date _____.
4) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date. _____ .
5) ☐ Notice of Informal Patent Application
6) ☐ Other: _____.

DETAILED ACTION

Election/Restrictions

1. Applicant's election with traverse of Group I in the reply filed on August 18, 2008 is acknowledged. The traversal is on the ground(s) that there is no evidence of record to show that the properties of the invention can be obtained by a different process. This is not found persuasive because the previous Action stated only that the claimed structure could be made by a different process; furthermore, it is unclear what claimed properties are referred to.

Applicant has also not elected a species, as required in the restriction.

2. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Marc A Patterson whose telephone number is 571-272-1497. The examiner can normally be reached on Mon - Fri 8:30 AM - 5:00 PM.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Rena Dye can be reached on 571-272-1498. 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 http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free).

/Marc A Patterson/
Primary Examiner, Art Unit 1794

Application Number	Application/Control No.	Applicant(s)/Patent under Reexamination	
	11/348,150	CLEVELAND ET AL.	
	Examiner	Art Unit	
	MARC A. PATTERSON	1794	

Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
Page 278 of 1179

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INTERNATIONAL PAPER

CINCINNATI TECHNOLOGY CENTER
6285 TRI-RIDGE BOULEVARD
LOVELAND OH 45140

T 513 248 6000
F 513 248 6455

CONFIDENTIAL FACSIMILE TRANSMISSION

To:	Commissioner for Patents
Fax:	571-273-8300
Tel:	
Date:	December 30, 2008
RE:	Restriction Requirement

Our File Ref.: FDS-029388-US
Your File Ref.: 11/348,150
No. of Pages: 4 (including this cover sheet)

☐ URGENT ☐ FOR YOUR INFORMATION

☐ CONFIRMATION COPY FOLLOWS BY:
☐ FIRST-CLASS U.S. MAIL
☐ OVERNIGHT MAIL
☐ INTERNATIONAL MAIL

☒ NO COPY FOLLOWS

In response to the Office Action dated 12/02/2008, please find the following checked items:

☐ Petition for Extension of Time Under 37 C.F.R. 1.136(a), Form PTO/SB/22, and one (1) copy thereof attached thereto, _____ sheet(s);

☒ Response to Restriction Requirement, 2 sheet(s);

☐ Fee Transmittal, Form PTO/SB/17, _____ sheet(s); and,

☐ Information Disclosure Statement by Applicant(s), _____ sheet(s), to which Form PTO/SB/08, _____ sheet(s), has been attached along with a copy of each reference(s) cited thereon.

Thank you.

Certificate of Transmission Under 37 C.F.R. 1.8

The undersigned hereby certifies that a true and accurate copy of the items checked above are being transmitted to the Honorable Commissioner for Patents, by facsimile transmission to the facsimile number indicated above, on this the 30th *day of* December, 20 08.

Michele L McRoy

Michele L. McRoy

This facsimile transmission may contain confidential and/or legally privileged information from the law department of International Paper Company which is intended only for the use of the individual(s) named above. If you are not the intended recipient, you are hereby notified that any disclosure, copying or distribution of this facsimile transmission is strictly prohibited. If you have received this transmission in error, please notify us immediately by phoning (513) 248-6207 so that we can arrange for the return of the documents. Thank you.

INTERNATIONAL PAPER

MATTHEW M. ESLAMI, PE
PATENT AGENT
INTELLECTUAL PROPERTY

CINCINNATI TECHNOLOGY CENTER
6285 TRI-RIDGE BOULEVARD
LOVELAND OH 45140

T 513 248 6193
F 513 248 6455
matthew.eslami@ipaper.com

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DEC 30 2008

SENT BY ☒ FACSIMILE TO (571-273-8300)
☐ FIRST CLASS MAIL TO THE ADDRESS BELOW

December 30, 2008

Commissioner for Patents
Post Office Box 1450
Alexandria, Virginia 22313-1450

RE: RESPONSE TO RESTRICTION REQUIREMENT

Applicant	:	Christopher S. Cleveland
Serial No.	:	11/348,150
Filed on	:	02/06/2006
Title	:	Biodegradable Paper-Based Laminate With Oxygen And Moisture Barrier Properties And Method.....
Our Ref.	:	FDS-029388-US

Dear Commissioner:

Enclosed herewith for filing, Applicant(s) respectfully submit(s) the following checked items:

☐ Petition for Extension of Time Under 37 C.F.R. 1.136(a), Form PTO/SB/22, and one (1) copy thereof attached thereto, _____ sheet(s);

☒ Response to Restriction Requirement, __2__ sheet(s);

☐ Fee Transmittal, Form PTO/SB/17, _____ sheet(s);

☐ Information Disclosure Statement by Applicant(s), _____ sheet(s), to which Form PTO/SB/08, _____ sheet(s), has been attached along with a copy of each reference(s) cited thereon; and,

☐ a postage-prepaid, return-receipt postcard with a listing of the above-checked items.

Please stamp the enclosed postcard and return same to me to indicate your receipt of the above-listed items. Please feel free to contact me if you have any questions concerning the above or the enclosed.

With kindest regards,

Matthew M. Eslami
Agent for Applicant(s)

Enclosures
MME/mlm

CERTIFICATE OF MAILING OR TRANSMISSION UNDER 37 C.F.R. § 1.8

The undersigned hereby certifies that a true and accurate copy of the documents checked hereon, together with all attachments referred-to herein, is being transmitted to the Honorable Commissioner for Patents, either by first-class mail, postage prepaid, addressed to Commissioner for Patents, Post Office Box 1450, Alexandria, Virginia 22313-1450, or by facsimile transmission to the facsimile number indicated hereon, on this the 30th day of December 20 08.

Michele L. McRoy

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DEC 30 2008

Docket No.: FDS-029388-US

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF:

CLEVELAND, et al. : GROUP ART UNIT: 1794

SERIAL NO: 11/348,150 : EXAMINER: PATTERSON, MARC A

FILED: February 06, 2006 :

FOR: BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOISTURE BARRIER PROPERTIES AND METHOD FOR MAKING BIODEGRADABLE PAPER-BASED LAMINATE

RESPONSE TO RESTRICTION REQUIREMENT

COMMISSIONER FOR PATENTS
UNITED STATES PATENT AND TRADEMARK OFFICE
PO BOX 1450
ALEXANDRIA VA 22313-1450

SIR:

Responsive to the Official Communication dated December 02, 2008, Applicants elect, without traverse, Specie (i), drawn to a laminate comprising a polymeric protectant layer, moisture protective layer and degradable tie layer (Claims 1-35, 38 and 41-43), for further prosecution.

PAGE 3/4 * RCVD AT 12/30/2008 2:52:16 PM [Eastern Standard Time] * SVR:USPTO-EFXRF-4/16 * DNIS:2738300 * CSID:513 248 6451 * DURATION (mm-ss):03-06

Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
Page 281 of 1179

The Applicants further submit that this application is in condition for examination on the merits and an early notification to that effect is earnestly solicited.

Date: December 30, 2008

By: _____

Matthew M. Eslami
Reg. No. 45,488
Patent Agent for Applicant
International Paper Company
6285 Tri-Ridge Boulevard
Loveland, Ohio 45140
Tel: (513) 248-6193
Fax: (513) 248-6455
e-mail: matt.eslami@ipaper.com

PAGE 4/4 * RCVD AT 12/30/2008 2:52:16 PM [Eastern Standard Time] * SVR:USPTO-EFXRF-4/16 * DNIS:2738300 * CSID:513 248 6451 * DURATION (mm-ss):03-06

Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
Page 282 of 1179

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
11/348,150	02/06/2006	Christopher S. Cleveland	FDS-029388-US	5681

1726 7590 03/19/2009

INTERNATIONAL PAPER COMPANY
6285 TRI-RIDGE BOULEVARD
LOVELAND, OH 45140

EXAMINER	
PATTERSON, MARC A	
ART UNIT	PAPER NUMBER
1794	

MAIL DATE	DELIVERY MODE
03/19/2009	PAPER

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.

Office Action Summary	Application No.	Applicant(s)	
	11/348,150	CLEVELAND ET AL.	
	Examiner	Art Unit	
	MARC A. PATTERSON	1794	

-- 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 MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, 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.
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- 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 _____.

2a)☐ This action is **FINAL**. 2b)☒ This action is non-final.

3)☐ 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

4)☒ Claim(s) *1-35,38,41-43 and 50-58* is/are pending in the application.

4a) Of the above claim(s) _____ is/are withdrawn from consideration.

5)☐ Claim(s) _____ is/are allowed.

6)☒ Claim(s) *1-35,38,41-43 and 50-58* is/are rejected.

7)☐ Claim(s) _____ is/are objected to.

8)☐ Claim(s) _____ are subject to restriction and/or election requirement.

Application Papers

9)☐ The specification is objected to by the Examiner.

10)☐ 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).

11)☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

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).

a)☐ All b)☐ Some * c)☐ None of:

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) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
3) ☒ Information Disclosure Statement(s) (PTO/SB/08) Paper No(s)/Mail Date *8/24/06;10/25/07*.
4) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date. _____ .
5) ☐ Notice of Informal Patent Application
6) ☐ Other: _____.

DETAILED ACTION

Claim Rejections – 35 USC § 102(b)

1. The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

> A person shall be entitled to a patent unless –
>
> (b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of application for patent in the United States.

2. Claims 1 – 5, 10, 12 – 13, 15, 22 – 23, 28, 30, 32, 41 and 43 are rejected under 35 U.S.C. 102(b) as being anticipated by Andersson et al. (U.S. Patent Publication No. 2005/0008800 A1).

With regard to Claims 1 – 2 and 12, Andersson et al discloses a laminate comprising substrate comprising a substrate (core of paperboard; paragraph 0017) a polymeric protectant layer on the outside surface for protecting the substrate from liquid and water vapor (paragraph 0031) and an oxygen barrier layer (aluminum foil; paragraph 0031) and product contact layer for preventing penetration by vapor, liquid and solid contents disposed on the oxygen barrier layer (coating '32' in Figure paragraph 0021) and a layer disposed on the inside surface of the substrate (adhesive; paragraph 0039) comprising polypropylene (paragraph 0039), therefore a moisture protective layer.

With regard to Claim 3, Andersson et al do not disclose bleaching of paperboard; unbleached paperboard is therefore disclosed by Andersson et al.

With regard to Claim 4, the polymeric protectant layer is polyethylene (paragraph 0036), therefore a print medium.

With regard Claim 5, the polymeric protectant layer comprises linear low density polyethylene (paragraph 0034), therefore a degradable polyethylene.

With regard Claims 10, 22, 23, 28 and 41, the moisture protective layer and product contact layer comprise linear low density polyethylene (paragraph 0034), therefore a degradable polyethylene.

With regard to Claims 13, 15, 30 and 32, the oxygen barrier comprises polyvinyl alcohol (paragraph 0033).

With regard to Claim 43, Andersson et al disclose a drinking cup (carton; paragraph 0018).

Claim Rejections – 35 USC § 103(a)

3. The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

> (a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negatived by the manner in which the invention was made.

4. Claims 6 – 9, 11, 14, 16 – 21, 24 – 27, 29, 31, 42 and 57 – 58 are rejected under 35 U.S.C. 103(a) as being unpatentable over Andersson et al. (U.S. Patent Publication No. 2005/0008800 A1).

Andersson et al disclose a laminate as discussed above. With regard to Claims 6 – 9, 11, 14, 16 – 21, 24 – 27, 29, 31, 42 and 57 – 58, Andersson et al fail to disclose a polymeric protectant layer, moisture protective layer and product contact layer having a coat weight of from five to twenty pounds per three thousand feet and degradable polyolefin comprising a catalyst comprising iron or ethylene – co – maleic anhydride. However, Andersson et al discloses layers comprised in packaging (paragraph 0020), and polyethylene, and a laminate comprising an

adhesive layer, therefore a tie layer, as discussed above. It therefore would have been obvious for one of ordinary skill in the art to have provided for metallocene polyethylene, depending on the desired method of synthesis, and to have provided for ethylene – co – maleic anhydride, depending on desired adhesive properties, and to have provided for additional tie layers and a desired coat weight of polymeric protectant layer, moisture protective layer and product contact layer depending on the desired weight of the package.

5. Claims 33 – 35, 38 and 50 – 56 are rejected under 35 U.S.C. 103(a) as being unpatentable over Andersson et al. (U.S. Patent Publication No. 2005/0008800 A1) in view of Gen (U.S. Patent No. 6,949,254 B2).

Andersson et al disclose a laminate as discussed above. The laminate comprises polyester (paragraph 0024). With regard to Claims 33 – 35, 38, 51 – 53 and 55, Andersson et al fail to disclose a polymeric protectant layer, moisture protective layer and product contact layer comprising polylactic acid, vitamin E, zeolite and starch.

Gen teaches a polyester comprising polylactic acid (column 3, lines 44 - 45), vitamin E, (column 2, lines 42 – 44), zeolite (column 3, lines 10 – 14) and starch (column 1, lines 30 – 31) for the purpose of using a polymer having good resistance to thermal decomposition. One of ordinary skill in the art would therefore have recognized the advantage of providing for the polylactic acid, vitamin E, zeolite and starch of Gen in Andersson et al, which comprises a polyester, depending on the desired resistance to thermal decomposition of the end product.

It therefore would have been obvious for one of ordinary skill in the art at the time Applicant's invention was made to have provided for polylactic acid, vitamin E, zeolite and

starch in Andersson et al in order to obtain good resistance to thermal decomposition as taught by Gen.

With regard to Claims 50 and 56, Gen fails to teach calcium carbonate and zeolite in the amount at most five weight percent. However, Gen teaches the use of any inorganic compound (column 3, lines 10 - 14) for the purpose of obtaining a complex (column 3, lines 10 -14). It therefore would have been obvious for one of ordinary skill in the art to have provided for calcium carbonate, as calcium carbonate constitutes an inorganic compound, or to have provided for a desired amount of inorganic compound depending on the desired chemical composition of the end product.

With regard to Claim 54, Gen teaches vitamin E in the amount of five percent by weight (column 2, lines 62 - 65).

6. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Marc A Patterson whose telephone number is 571-272-1497. The examiner can normally be reached on Mon - Fri 8:30 AM - 5:00 PM.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Rena Dye can be reached on 571-272-1498. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

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/Marc A Patterson/
Primary Examiner, Art Unit 1794

Notice of References Cited	Application/Control No. 11/348,150	Applicant(s)/Patent Under Reexamination CLEVELAND ET AL.	
	Examiner MARC A. PATTERSON	Art Unit 1794	Page 1 of 1

U.S. PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Name	Classification
*	A	US-2005/0008800	01-2005	Andersson et al.	428/034.2
*	B	US-6,949,254	09-2005	Gen, Shokyu	424/426
	C	US-			
	D	US-			
	E	US-			
	F	US-			
	G	US-			
	H	US-			
	I	US-			
	J	US-			
	K	US-			
	L	US-			
	M	US-			

FOREIGN PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Country	Name	Classification
	N					
	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.

OIPE IAP60
AUG 24 2006
PATENT & TRADEMARK OFFICE

JFW

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
WASHINGTON, D.C., UNITED STATES OF AMERICA

In re Application of:

Applicant(s): CLEVELAND, et al.

Serial No.: 11/348,150

Filing Date: FEBRUARY 6, 2006

Entitled: BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN .....

Examiner:

Our File Ref.: FDS-029388-US

Group Art Unit: 1772

INFORMATION DISCLOSURE STATEMENT
SUBMITTED PERSUANT TO 37 C.F.R. 1.97(b)(1)

Commissioner for Patents
United States Patent and Trademark Office
Post Office Box 1450
Alexandria, Virginia 22313-1450

Dear Commissioner:

In accordance with the duty of disclosure under 37 C.F.R. 1.56, Applicant(s) hereby notifies the U.S. Patent and Trademark Office of certain documents which the Examiner might deem relevant to the issue of patentability of the claims pending in the above-referenced patent application. Enclosed herewith please find Form PTO/SB/08, Information Disclosure Statement by Applicant, 3 sheet(s), and a copy of each reference cited thereon.

This Information Disclosure Statement is filed before the mailing date of a first Office Action on the merits. As such, neither a fee under 37 C.F.R. 1.17(p) nor a statement under 37 C.F.R. 1.97(e) is required; Applicant(s) respectfully requests pursuant to 37 C.F.R. 1.97(b)(3) that the Examiner considers each of the references disclosed herewith.

The submission of the documents enclosed herewith is not intended as an admission that such documents constitute prior art under Title 35 of the United States Code, as against the claims of the above-referenced patent application. Applicant(s) does not waive any right to take

action that would be appropriate to antedate or otherwise overcome the use of any document submitted herewith as prior art under Title 35 of the United States Code.

Applicant(s) respectfully requests that the Examiner initial each of the references listed on the attached Form PTO/SB/08 and return same to the undersigned attorney in accordance with MPEP 609.

Should the Examiner have any questions relating to the submission of the within Information Disclosure Statement or relating to the copies submitted therewith, Applicant(s) respectfully request that the Examiner contact the undersigned counsel, who can be reached at the telephone numbers listed below.

Respectfully submitted,

Date: August 21, 2006

Daniel C. Stelter, Reg. 40,830
Counsel for Applicant
International Paper Company
6285 Tri-Ridge Boulevard
Loveland, Ohio 45140
513.248.6000 telephone
513.248.6455 facsimile

Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
Page 292 of 1179

OIPE IAP60
AUG 24 2006
PATENT & TRADEMARK OFFICE

INTERNATIONAL PAPER

DANIEL C. STELTER
SENIOR COUNSEL
INTELLECTUAL PROPERTY

CINCINNATI TECHNOLOGY CENTER
6285 TRI-RIDGE BOULEVARD
LOVELAND OH 45140

T 513 248 6208
F 513 248 6455
daniel.stelter@ipaper.com

August 21, 2006

Commissioner for Patents
Post Office Box 1450
Alexandria, Virginia 22313-1450

RE: SUBMISSION OF INFORMATION DISCLOSURE STATEMENT

Applicant	:	CLEVELAND, et al.
Serial No.	:	11/348,150
Filed on	:	FEBRUARY 6, 2006
Title	:	BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN.....
Our Ref.	:	FDS-029388-US

Dear Commissioner:

Enclosed herewith for filing, Applicant(s) respectfully submit(s) the following checked items:

☒ Information Disclosure Statement by Applicant(s), 2 sheet(s), to which Form PTO/SB/08, 3 sheet(s), has been attached along with a copy of each reference(s) cited thereon;

☐ Supplemental Information Disclosure Statement by Applicant(s), _____ sheet(s), to which Form PTO/SB/08, _____ sheet(s), has been attached along with a copy of each reference(s) cited thereon; and,

☒ a postage-prepaid, return-receipt postcard with a listing of the above-checked items.

Please stamp the enclosed postcard and return same to me to indicate your receipt of the above-listed items. Please feel free to contact me if you have any questions concerning the above or the enclosed.

With kindest regards,

Daniel C. Stelter
Attorney for Applicant(s)

Enclosures
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CERTIFICATE OF MAILING OR TRANSMISSION UNDER 37 C.F.R. § 1.8

The undersigned hereby certifies that a true and accurate copy of the documents checked hereon, together with all attachments referred-to herein, is being transmitted to the Honorable Commissioner for Patents, either by first-class mail, postage prepaid, addressed to Commissioner for Patents, Post Office Box 1450, Alexandria, Virginia 22313-1450, or by facsimile transmission to the facsimile number indicated hereon, on this the 21st day of August 20 06.

Jane A. Tomlinson

CIPE IAP60 AUG 24 2006 PATENT & TRADEMARK OFFICE

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Sheet 1 of 3

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Filing Date	FEBRUARY 6, 2006
First Named Inventor	CLEVELAND, et al.
Art Unit	~~1772~~ 1794
Examiner Name	M. Patterson
Attorney Docket Number	FDS-029388-US

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/M.P./	N	US- 5,854,304	12-29-1998		
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/M.P./	P	US- 6,033,747	03-07-2000		
/M.P./	Q	US- 6,040,063	03-21-2000		
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/M.P./	F2	JP2003-276144	09-30-2003			
/M.P./	F3	JP2003-41142	02-13-2003			

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/M.P./	V	US- 6,235,815	05-22-2001		
/M.P./	W	US- 6,245,437	06-12-2001		
/M.P./	X	US- 6,248,862	06-19-2001		
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/M.P./	I	US- 5,602,227	02-11-1997	Isao Noda	
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Substitute for form 1449/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Sheet 4 of 4

Complete if Known	
Application Number	11/348,150
Filing Date	FEBRUARY 6, 2006
First Named Inventor	CLEVELAND, et al.
Art Unit	~~1772~~ 1794
Examiner Name	M. Patterson
Attorney Docket Number	FDS-029388-US

U. S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number Number-Kind Code[2] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
/M.P./	A	US- WO/06/031568	03-23-2006	Cleveland, et al	
/M.P./	B	US- WO/01/61043	08-23-2001	Jian-Bing Fan	
/M.P./	C	US- WO/04/082936	09-30-2004	Rieghard, et al	
		US-			
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FOREIGN PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Foreign Patent Document Country Code[3]-Number[4]-Kind Code[5] (if known)	Publication Date MM-DD-YYYY	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages Or Relevant Figures Appear	T[6]
/M.P./	F1	EP 0629975	12-21-1994			
/M.P./	F2	DE 19954404	11-12-1999			
/M.P./		WO 01/36518	05-25-01			
/M.P./	F3	JP 04299131	10-22-1992			
		~~English~~				

Examiner Signature	/Marc Patterson/	Date Considered	03/16/2009

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to applicant. [1] Applicant's unique citation designation number (optional). [2] See Kinds Codes of USPTO Patent Documents at www.uspto.gov or MPEP 901.04. [3] Enter Office that issued the document, by the two-letter code (WIPO Standard ST.3). [4] For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. [5] Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. [6] Applicant is to place a check mark here if English language Translation is attached.

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 2 hours 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, 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 (1-800-786-9199) and select option 2.

Docket No.: FDS-029388-US

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF:

CLEVELAND, et al.	:	GROUP ART UNIT: 1794
SERIAL NO: 11/348,150	:	EXAMINER: PATTERSON, MARC A
FILED: February 06, 2006	:	

FOR: BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOISTURE BARRIER PROPERTIES AND METHOD FOR MAKING BIODEGRADABLE PAPER-BASED LAMINATE

AMENDMENT

COMMISSIONER FOR PATENTS
UNITED STATES PATENT AND TRADEMARK OFFICE
PO BOX 1450
ALEXANDRIA VA 22313-1450

Dear Commissioner:

Responsive to the Office Action mailed **March 19, 2009**, please accept the following Amendment and Remarks in response thereto.

Amendments to the claims begin on page **2** of this paper.

Remarks/Arguments begin on page **5** of this paper.

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Amendments to the Claims:

Listing of claims will replace all prior versions, and listings, of claims in the application:

Listing of Claims:

Claims 1-58. (Canceled)

Claims 59-73. (Canceled)

74. (New) A laminate for forming moisture barriers, comprising:

a substrate having an interior surface for contacting products; and

a moisture protectant layer disposed on said interior surface of said substrate and comprising at least one degradable polymer selected from the group consisting of a degradable polyolefin, a polyhydroxyalkanoate, polylactic acid, and a degradable polyester.

75. (New) The laminate according to claim 74, wherein the moisture protectant layer has a thickness of from 5 to 30 lbs/3000 ft^2.

76. (New) The laminate according to Claim 74, further comprising a second moisture protectant layer disposed on an exterior surface of said substrate and comprising at least one degradable polymer selected from the group consisting of a degradable polyolefin, a polyhydroxyalkanoate, polylactic acid, and a degradable polyester.

77. (New) The laminate according to Claim 76, wherein the second moisture protectant layer further comprises at least one filler.

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78. (New) The laminate according to Claim 76, wherein the second moisture protectant layer further comprises at least one member selected from the group consisting of calcium carbonate, starch, clay kaolin, talc, cellulose fibers, and diatomaceous earth.

79. (New) The laminate according to claim 76, wherein the second moisture protectant layer has a thickness of from 5 to 30 lbs/3000 ft^2.

80. (New) The laminate according to Claim 76, wherein the second moisture protectant layer further comprises at least one member selected from the group consisting of calcium carbonate and starch.

81. (New) The laminate according to Claim 74, wherein the moisture protectant layer further comprises at least one filler.

82. (New) The laminate according to Claim 74, wherein the moisture protectant layer further comprises at least one member selected from the group consisting of calcium carbonate, starch, clay kaolin, talc, cellulose fibers, and diatomaceous earth.

83. (New) The laminate according to Claim 74, wherein the moisture protectant layer further comprises at least one member selected from the group consisting of calcium carbonate and starch.

84. (New) The laminate according to Claim 74, wherein the substrate comprises paperboard; and the moisture protectant layer has a thickness of from 5 to 30 lbs/3000 ft^2 and comprises

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polylactic acid and at least one member selected from the group consisting of calcium carbonate and starch.

85. (New) A cup, comprising the laminate according to Claim 84.

86. (New) A cup, comprising the laminate according to Claim 74.

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Remarks/Arguments

By this Amendment, claims 1-58 are canceled. Claims 59-73 are withdrawn. New claims 74-86 are added. Support for new claims 74-86 can be found through out the specification, particularly, pages 6-9. Reconsideration in view of the above amendment is respectfully requested.

1. Interview Acknowledgment

Applicants thank Examiner Patterson for the courtesies extended to Applicant's representative during June 16, 2009 telephonic interview. During the interview, a draft of new claims 74-86 was discussed and it was understood that the new claims 74-86 define patenable subject matter over the cited references in the Office Action. It was decided that the Applicants submit claims 74-86 to replace originally submitted claims 1-58. The substance of the interview is summarized in the following remarks.

2. Rejection of Respective Claims 1-5, 10, 12-13, 15, 22-23, 28, 30, 32, 41, and 43 under 35 U.S.C. § 102 (b) as being anticipated by Andersson et al (U.S. Patent Publication No. 2005/0008800 A1).

Since claims 1-58 are canceled, then rejections of these claims are now rendered moot. However, Applicants submit new claims 74-86 that define patenable subject matter over the cited references in the Office Action. In the present invention, the polymeric protectant layer comprises linear low density polyethylene (Please see Spec., Paragraph 0034), therefore a degradable polyethylene. High density, low density, and linear low density describe different types of polyethylene. Each is defined by the amount and type of branching from the polymer backbone. As a result, each has different levels of crystallinity.

In the present invention, the inventors claim the use of a degradable polyethylene or

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polypropylene. Regular polyethylene is not considered to be degradable. Degradability is defined as a >75-80% loss in molecular weight (MWT) in a relatively short amount of time (<9 months) due to exposure to oxygen, water, and microbes in the environment. This process is the same as the degradation of other organic materials like food waste. Normal polyethylene is not considered to be degradable because it might take tens to hundreds of years to achieve the >75% loss in molecular weight. Standards and definitions for degradability are contained in ASTM D6400, ASTM D6868, and ASTM D5511. More specifically, degradable polyethylene is defined in ASTM D3826.

The Andersson's patent is directed to the use of normal, non-degradable polyolefins such as polyethylene, linear low density polyethylene, and polypropylene. These are general classes of polyolefins. The present invention uses the degradable versions of these polymers. Applicants respectfully requests withdrawal of this rejection.

3. Rejection of Claims 6-9, 11, 14, 16-21, 24-27, 29, 31, 42 and 57-58 under 35 U.S.C. § 103 (a) as being unpatentable over Andersson et al (U.S. Patent Publication No. 2005/0008800 A1).

Since claims 1-58 are canceled, then rejection of these claims are now rendered moot. Applicants respectfully requests withdrawal of this rejection.

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Clearwater Paper Corp. v. Graphic Packaging International LLC
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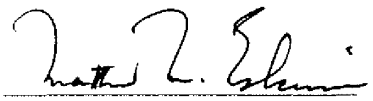
CONCLUSION

Based upon the foregoing, Applicants respectfully submit that claims 74-86 particularly define and patentably distinguish the present invention over the cited references. Accordingly, reconsideration of the rejection and allowance of all claims are earnestly requested. However, should the Examiner have any remaining questions and the attending to of which would expedite such action, the Examiner is invited to contact the undersigned at the telephone number listed below.

No month extension of time is believed to be required. However, the Commissioner is authorized to charge any fees associated with this or any other communication, or credit any over payment, to Deposit Account No. 09-0525.

Respectfully Submitted,

Date: June 19, 2009

By: 

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Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
Page 307 of 1179

Electronic Acknowledgement Receipt	
EFS ID:	5554127
Application Number:	11348150
International Application Number:	
Confirmation Number:	5681
Title of Invention:	Biodegradable paper-based laminate with oxygen and moisture barrier properties and method for making biodegradable paper-based laminate
First Named Inventor/Applicant Name:	Christopher S. Cleveland
Customer Number:	01726
Filer:	Matthew M. Eslami/Michele McRoy
Filer Authorized By:	Matthew M. Eslami
Attorney Docket Number:	FDS-029388-US
Receipt Date:	19-JUN-2009
Filing Date:	06-FEB-2006
Time Stamp:	16:48:40
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		090619_rsptoofficeaction_fds029388us.pdf	93609 c1aaf89cef8af9f4cd1ae1c6dba4df4c61371a09	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:			
	Total Files Size (in bytes):	93609	

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 (07-06)
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PATENT APPLICATION FEE DETERMINATION RECORD
Substitute for Form PTO-875

Application or Docket Number	Filing Date	☐ To be Mailed
11/348,150	02/06/2006	

APPLICATION AS FILED – PART I

	(Column 1)	(Column 2)	SMALL ENTITY ☐ RATE ($)	SMALL ENTITY ☐ FEE ($)	OR	OTHER THAN SMALL ENTITY RATE ($)	OTHER THAN SMALL ENTITY FEE ($)
FOR	NUMBER FILED	NUMBER EXTRA					
☐ BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A	N/A			N/A	
☐ SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A	N/A			N/A	
☐ EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A	N/A			N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	minus 20 =	*	X $ =		OR	X $ =	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	minus 3 =	*	X $ =			X $ =	
☐ APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $250 ($125 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))							
			TOTAL			TOTAL	

* If the difference in column 1 is less than zero, enter "0" in column 2.

APPLICATION AS AMENDED – PART II

AMENDMENT	06/19/2009	(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 ($)	OTHER THAN SMALL ENTITY ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	* 13	Minus	** 73	= 0	X $ =		OR	X $52=	0
	Independent (37 CFR 1.16(h))	* 1	Minus	***13	= 0	X $ =		OR	X $220=	0
	☐ 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	0

AMENDMENT		(Column 1) CLAIMS REMAINING AFTER AMENDMENT		(Column 2) HIGHEST NUMBER PREVIOUSLY PAID FOR	(Column 3) PRESENT EXTRA	RATE ($)	ADDITIONAL FEE ($)		RATE ($)	ADDITIONAL FEE ($)
	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 number found in the appropriate box in column 1.

Legal Instrument Examiner:
/BRENDA WEBB/

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.**
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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
11/348,150	02/06/2006	Christopher S. Cleveland	FDS-029388-US	5681

1726 7590 10/28/2009

INTERNATIONAL PAPER COMPANY
6285 TRI-RIDGE BOULEVARD
LOVELAND, OH 45140

EXAMINER	
PATTERSON, MARC A	
ART UNIT	PAPER NUMBER
1794	

MAIL DATE	DELIVERY MODE
10/28/2009	PAPER

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.

Office Action Summary	Application No.	Applicant(s)	
	11/348,150	CLEVELAND ET AL.	
	Examiner	Art Unit	
	MARC A. PATTERSON	1794	

-- 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 MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, 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 *19 June 2009*.

2a)☐ This action is **FINAL**. 2b)☒ This action is non-final.

3)☐ 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

4)☒ Claim(s) *74-86* is/are pending in the application.

4a) Of the above claim(s) _____ is/are withdrawn from consideration.

5)☐ Claim(s) _____ is/are allowed.

6)☒ Claim(s) *74-86* is/are rejected.

7)☐ Claim(s) _____ is/are objected to.

8)☐ Claim(s) _____ are subject to restriction and/or election requirement.

Application Papers

9)☐ The specification is objected to by the Examiner.

10)☐ 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).

11)☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

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).

a)☐ All b)☐ Some * c)☐ None of:

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) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
3) ☐ Information Disclosure Statement(s) (PTO/SB/08) Paper No(s)/Mail Date _____.
4) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date. _____ .
5) ☐ Notice of Informal Patent Application
6) ☐ Other: _____.

DETAILED ACTION

WITHDRAWN REJECTIONS

1. The 35 U.S.C. 102(b) rejection of Claims 74, 76 and 85 – 86 as being anticipated by Andersson et al. (U.S. Patent Publication No. 2005/0008800 A1), of record on page 2 of the previous Action, is withdrawn.

NEW REJECTIONS

Claim Rejections – 35 USC § 103(a)

2. The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

> (a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negatived by the manner in which the invention was made.

3. Claims 74 – 76, 79 and 84 – 86 are rejected under 35 U.S.C. 103(a) as being unpatentable over Andersson et al. (U.S. Patent Publication No. 2005/0008800 11) in view of Reich et al (U.S. Patent No. 3,984,940).

With regard to Claim 74, Andersson et al discloses a laminate comprising substrate comprising a substrate (core of paperboard; paragraph 0017) and an interior moisture protective layer (coating '32' in Figure; paragraph 0021) comprising polypropylene (paragraph 0036). Andersson et al fail to disclose a polypropylene that is degradable.

Reich et al teach discloses a polypropylene (column 11, lines 54 – 61) that is degradable, for the purpose of obtaining a film that can be degraded by solar radiation (column 12, lines 10 - 14). It therefore would have been obvious for one of ordinary skill in the art at the time

Applicant's invention was made to have provided for a degradable polypropylene in order to provide a film that degrades by solar radiation as taught by Andersson et al.

With regard to Claims 75, 79 and 84, Andersson et al fail to disclose a polymeric protectant layer, moisture protective layer and product contact layer having a thickness of from five to thirty pounds per three thousand feet. However, Andersson et al discloses layers comprised in packaging (paragraph 0020), and polyethylene, and a laminate, as discussed above. It therefore would have been obvious for one of ordinary skill in the art to have provided for a desired coat weight of polymeric protectant layer, moisture protective layer and product contact layer depending on the desired weight of the package.

With regard Claim 76, the moisture protective layer and product contact layer comprise linear low density polyethylene (paragraph 0034), therefore a degradable polyethylene.

With regard to Claims 85 – 86, Andersson et al disclose a drinking cup (carton; paragraph 0018).

4. Claims 77 – 78 and 80 - 83 are rejected under 35 U.S.C. 103(a) as being unpatentable over Andersson et al. (U.S. Patent Publication No. 2005/0008800 11) in view of Reich et al (U.S. Patent No. 3,984,940) and further in view of Gen (U.S. Patent No. 6,949,254 B2).

Andersson et al and Reich et al disclose a laminate as discussed above. The laminate comprises polyester (paragraph 0024). With regard to Claims 77 – 78 and 80 - 83, Andersson et al and Reich et al fail to disclose polylactic acid and a filler comprising starch.

Gen teaches a polyester comprising polylactic acid (column 3, lines 44 - 45) and starch (column 1, lines 30 - 31) for the purpose of using a polymer having good resistance to thermal

decomposition. One of ordinary skill in the art would therefore have recognized the advantage of providing for the polylactic acid and starch of Gen in Andersson et al and Reich et al, which comprises a polyester, depending on the desired resistance to thermal decomposition of the end product.

It therefore would have been obvious for one of ordinary skill in the art at the time Applicant's invention was made to have provided for polylactic acid and starch in Andersson et al and Reich et al in order to obtain good resistance to thermal decomposition as taught by Gen.

ANSWERS TO APPLICANT'S ARGUMENTS

5. Applicant's arguments regarding the rejections of the previous Action have been considered and have been found to be persuasive. The new rejections above are directed to new amended Claims 74 – 86.

6. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Marc A Patterson whose telephone number is 571-272-1497. The examiner can normally be reached on Mon - Fri 8:30 AM - 5:00 PM.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Rena Dye can be reached on 571-272-1498. 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 http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free).

/Marc A Patterson/
Primary Examiner, Art Unit 1794

Notice of References Cited	Application/Control No. 11/348,150	Applicant(s)/Patent Under Reexamination CLEVELAND ET AL.	
	Examiner MARC A. PATTERSON	Art Unit 1794	Page 1 of 1

U.S. PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Name	Classification
*	A	US-3,833,401	09-1974	Ingram, Alvin R.	427/256
*	B	US-3,984,940	10-1976	Reich et al.	47/9
	C	US-			
	D	US-			
	E	US-			
	F	US-			
	G	US-			
	H	US-			
	I	US-			
	J	US-			
	K	US-			
	L	US-			
	M	US-			

FOREIGN PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Country	Name	Classification
	N					
	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.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
United States Patent and Trademark Office
Address: COMMISSIONER FOR PATENTS
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www.uspto.gov

APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
11/348,150	02/06/2006	Christopher S. Cleveland	FDS-029388-US	5681

1726 7590 10/30/2009

INTERNATIONAL PAPER COMPANY
6285 TRI-RIDGE BOULEVARD
LOVELAND, OH 45140

EXAMINER	
PATTERSON, MARC A	
ART UNIT	PAPER NUMBER
1794	

MAIL DATE	DELIVERY MODE
10/30/2009	PAPER

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.

Interview Summary	Application No.	Applicant(s)	
	11/348,150	CLEVELAND ET AL.	
	Examiner	Art Unit	
	MARC A. PATTERSON	1794	

All participants (applicant, applicant's representative, PTO personnel):

(1) *Marc Patterson*. (3)_____.

(2) *Matt Eslami*. (4)_____.

Date of Interview: *16 June 2009*.

Type: a)☒ Telephonic b)☐ Video Conference
c)☐ Personal [copy given to: 1)☐ applicant 2)☐ applicant's representative]

Exhibit shown or demonstration conducted: d)☐ Yes e)☒ No.
If Yes, brief description: _____.

Claim(s) discussed: *all of the record*.

Identification of prior art discussed: *all of the record*.

Agreement with respect to the claims f)☐ was reached. g)☒ was not reached. h)☐ N/A.

Substance of Interview including description of the general nature of what was agreed to if an agreement was reached, or any other comments: *The claimed invention vs the prior art of record, specifically if the claimed layers are disclosed by the prior art of record*.

(A fuller description, if necessary, and a copy of the amendments which the examiner agreed would render the claims allowable, if available, must be attached. Also, where no copy of the amendments that would render the claims allowable is available, a summary thereof must be attached.)

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. See Summary of Record of Interview requirements on reverse side or on attached sheet.

/Marc A Patterson/ Primary Examiner, Art Unit 1794	

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Summary of Record of Interview Requirements

Manual of Patent Examining Procedure (MPEP), Section 713.04, Substance of Interview Must be Made of Record
A complete written statement as to the substance of any face-to-face, video conference, or telephone interview with regard to an application must be made of record in the application whether or not an agreement with the examiner was reached at the interview.

Title 37 Code of Federal Regulations (CFR) § 1.133 Interviews
Paragraph (b)

In every instance where reconsideration is requested in view of an interview with an examiner, a complete written statement of the reasons presented at the interview as warranting favorable action must be filed by the applicant. An interview does not remove the necessity for reply to Office action as specified in §§ 1.111, 1.135. (35 U.S.C. 132)

37 CFR §1.2 Business to be transacted in writing.
All business with the Patent or Trademark Office should be transacted in writing. The personal attendance of applicants or their attorneys or agents at the Patent and Trademark Office is unnecessary. The action of the Patent and Trademark Office will be based exclusively on the written record in the Office. No attention will be paid to any alleged oral promise, stipulation, or understanding in relation to which there is disagreement or doubt.

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The action of the Patent and Trademark Office cannot be based exclusively on the written record in the Office if that record is itself incomplete through the failure to record the substance of interviews.

It is the responsibility of the applicant or the attorney or agent to make the substance of an interview of record in the application file, unless the examiner indicates he or she will do so. It is the examiner's responsibility to see that such a record is made and to correct material inaccuracies which bear directly on the question of patentability.

Examiners must complete an Interview Summary Form for each interview held where a matter of substance has been discussed during the interview by checking the appropriate boxes and filling in the blanks. Discussions regarding only procedural matters, directed solely to restriction requirements for which interview recordation is otherwise provided for in Section 812.01 of the Manual of Patent Examining Procedure, or pointing out typographical errors or unreadable script in Office actions or the like, are excluded from the interview recordation procedures below. Where the substance of an interview is completely recorded in an Examiners Amendment, no separate Interview Summary Record is required.

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- Application Number (Series Code and Serial Number)
- Name of applicant
- Name of examiner
- Date of interview
- Type of interview (telephonic, video-conference, or personal)
- Name of participant(s) (applicant, attorney or agent, examiner, other PTO personnel, etc.)
- An indication whether or not an exhibit was shown or a demonstration conducted
- An identification of the specific prior art discussed
- An indication whether an agreement was reached and if so, a description of the general nature of the agreement (may be by attachment of a copy of amendments or claims agreed as being allowable). Note: Agreement as to allowability is tentative and does not restrict further action by the examiner to the contrary.
- The signature of the examiner who conducted the interview (if Form is not an attachment to a signed Office action)

It is desirable that the examiner orally remind the applicant of his or her obligation to record the substance of the interview of each case. It should be noted, however, that the Interview Summary Form will not normally be considered a complete and proper recordation of the interview unless it includes, or is supplemented by the applicant or the examiner to include, all of the applicable items required below concerning the substance of the interview.

A complete and proper recordation of the substance of any interview should include at least the following applicable items:

1) A brief description of the nature of any exhibit shown or any demonstration conducted,
2) an identification of the claims discussed,
3) an identification of the specific prior art discussed,
4) an identification of the principal proposed amendments of a substantive nature discussed, unless these are already described on the Interview Summary Form completed by the Examiner,
5) a brief identification of the general thrust of the principal arguments presented to the examiner,
   (The identification of arguments need not be lengthy or elaborate. A verbatim or highly detailed description of the arguments is not required. The identification of the arguments is sufficient if the general nature or thrust of the principal arguments made to the examiner can be understood in the context of the application file. Of course, the applicant may desire to emphasize and fully describe those arguments which he or she feels were or might be persuasive to the examiner.)
6) a general indication of any other pertinent matters discussed, and
7) if appropriate, the general results or outcome of the interview unless already described in the Interview Summary Form completed by the examiner.

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Examiner to Check for Accuracy

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11/348,150	02/06/2006	Christopher S. Cleveland	FDS-029388-US	5681

1726 7590 01/27/2010

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Interview Summary	Application No.	Applicant(s)	
	11/348,150	CLEVELAND ET AL.	
	Examiner	Art Unit	
	MARC A. PATTERSON	1794	

All participants (applicant, applicant's representative, PTO personnel):

(1) *Marc Patterson*. (3)*Mark Fagan*.

(2) *Matt Eslami*. (4)_____.

Date of Interview: *22 January 2010*.

Type: a)☒ Telephonic b)☐ Video Conference
c)☐ Personal [copy given to: 1)☐ applicant 2)☐ applicant's representative]

Exhibit shown or demonstration conducted: d)☐ Yes e)☒ No.
If Yes, brief description: _____.

Claim(s) discussed: *alll of the record*.

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Agreement with respect to the claims f)☐ was reached. g)☒ was not reached. h)☐ N/A.

Substance of Interview including description of the general nature of what was agreed to if an agreement was reached, or any other comments: *The claimed invention vs the prior art of record, specifically whether the prior art of record discloses a degradable polymer as defined by Applicant's invention*.

(A fuller description, if necessary, and a copy of the amendments which the examiner agreed would render the claims allowable, if available, must be attached. Also, where no copy of the amendments that would render the claims allowable is available, a summary thereof must be attached.)

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/Marc A Patterson/ Primary Examiner, Art Unit 1794	

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Summary of Record of Interview Requirements

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Docket No.: FDS-029388-US

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF:

CLEVELAND, et al.	:	GROUP ART UNIT: 1794
SERIAL NO: 11/348,150	:	EXAMINER: PATTERSON, MARC A
FILED: February 06, 2006	:	CONFIRMATION #: 5681

FOR: BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOISTURE BARRIER PROPERTIES AND METHOD FOR MAKING BIODEGRADABLE PAPER-BASED LAMINATE

RESPONSE

COMMISSIONER FOR PATENTS
UNITED STATES PATENT AND TRADEMARK OFFICE
PO BOX 1450
ALEXANDRIA VA 22313-1450

Dear Commissioner:

Responsive to the Office Action mailed on **October 28, 2009**, please accept the following Amendment and Remarks in response thereto.

Amendments to the claims begin on page **2** of this paper.

Remarks/Arguments begin on page **5** of this paper.

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Amendments to the Claims:

Listing of claims will replace all prior versions, and listings, of claims in the application:

Listing of Claims:

Claims 1-58. (Canceled)

Claims 59-73. (Canceled)

74. (Previously presented) A laminate for forming moisture barriers, comprising:

a substrate having an interior surface for contacting products; and

a moisture protectant layer disposed on said interior surface of said substrate and comprising at least one degradable polymer selected from the group consisting of a degradable polyolefin, a polyhydroxyalkanoate, polylactic acid, and a degradable polyester.

75. (Previously presented) The laminate according to claim 74, wherein the moisture protectant layer has a thickness of from 5 to 30 lbs/3000 ft^2.

76. (Previously presented) The laminate according to Claim 74, further comprising a second moisture protectant layer disposed on an exterior surface of said substrate and comprising at least one degradable polymer selected from the group consisting of a degradable polyolefin, a polyhydroxyalkanoate, polylactic acid, and a degradable polyester.

77. (Previously presented) The laminate according to Claim 76, wherein the second moisture protectant layer further comprises at least one filler.

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78. (Previously presented) The laminate according to Claim 76, wherein the second moisture protectant layer further comprises at least one member selected from the group consisting of calcium carbonate, starch, clay kaolin, talc, cellulose fibers, and diatomaceous earth.

79. (Previously presented) The laminate according to claim 76, wherein the second moisture protectant layer has a thickness of from 5 to 30 lbs/3000 ft^2.

80. (Previously presented) The laminate according to Claim 76, wherein the second moisture protectant layer further comprises at least one member selected from the group consisting of calcium carbonate and starch.

81. (Previously presented) The laminate according to Claim 74, wherein the moisture protectant layer further comprises at least one filler.

82. (Previously presented) The laminate according to Claim 74, wherein the moisture protectant layer further comprises at least one member selected from the group consisting of calcium carbonate, starch, clay kaolin, talc, cellulose fibers, and diatomaceous earth.

83. (Previously presented) The laminate according to Claim 74, wherein the moisture protectant layer further comprises at least one member selected from the group consisting of calcium carbonate and starch.

84. (Previously presented) The laminate according to Claim 74, wherein the substrate

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comprises paperboard; and the moisture protectant layer has a thickness of from 5 to 30 lbs/3000 ft^2 and comprises polylactic acid and at least one member selected from the group consisting of calcium carbonate and starch.

85. (Previously presented) A cup, comprising the laminate according to Claim 84.

86. (Previously presented) A cup, comprising the laminate according to Claim 74.

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Remarks/Arguments

Claims 74-86 are currently pending. Favorable reconsideration is respectfully requested in light of the following remarks.

1. Interview Acknowledgment

Applicants thank Examiner Patterson for the courtesies extended to Applicant's representatives during January 22, 2010 telephonic interview. During the interview, claim 74 was discussed and it was understood that the term "degradable polyolefin" is defined within the scope of the description and therefore defines patenable subject matter over the cited references in the Office Action. The substance of the interview is summarized in the following remarks.

2. Claims 74-76, 79 and 84-86 are rejected under 35 U.S.C. § 103 (a) as being unpatentable over Andersson et al (U.S. Patent Publication No. 2005/0008800 A1) in view of Reich et al (USP #3,984,940)

Claim 74 of the present invention recites:

"a moisture protectant layer disposed on said interior surface of said substrate and comprising at least one degradable polymer selected from the group consisting of a degradable polyolefin, a polyhydroxyalkanoate, polylactic acid, and a degradable polyester." (Emphasis added)

The term degradable polyolefin is defined on paragraph [0031] as follows:

"A degradable polyolefin (dPE) is a blend of a transition metal carboxylate catalyst with polyethylene and polypropylene. Examples of transition metals used in the catalyst may include: iron, cadmium, cobalt, and manganese. Two commercial suppliers of this additive are Environmental Plastics, Inc. (EPI) and Willow Ridge Plastics, Inc."

Clearly, the present invention as defined by claim 74 is directed to a degradable polymer selected from the group consisting of a degradable polyolefin, a polyhydroxyalkanoate, polylactic

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acid, and degradable polyester. This aspect of the invention is neither taught nor suggested by **Andersson et al.** The **Andersson** patent is directed to the use of normal, non-degradable polyolefins such as polyethylene, linear low-density polyethylene, and polypropylene. These are general classes of polyolefins. The present invention uses the degradable versions of these polymers. **Andersson**'s patent is directed to a retortable packaging container that is intended principally for particularly perishable and oxygen gas-sensitive products such as juices and other foodstuffs. (Please see paragraph [0040]). Moreover, **Andersson**'s patent, as described in paragraph [0037], is directed to:

> "the liquid-tight coating 32 consists of a copolymer of propylene and ethylene which is sufficiently moisture and heat-resistant to withstand extreme moisture and temperature stresses which occur in a normal heat treatment for extending shelf-life which is carried out in a retort."(Emphasis added)

On the other hand, the present invention is directed to a laminate (Claim 74), particularly for a disposable cup as recited claims 85, and 86, made of degradable or compostable/renewable-resource-based materials that include polylactic acid (PLA) and polyhydroxyalkanoates. The moisture protectant layer of claim 74 comprises linear low density polyethylene (Please see Spec., Paragraph 0034), therefore a degradable polyethylene. High density, low density, and linear low density describe different types of polyethylene. Each is defined by the amount and type of branching from the polymer backbone. The inventors of the present invention claim the use of a degradable polyethylene or polypropylene. Regular polyethylene is not considered to be degradable. Therefore, the **Andersson et al.** patent neither teaches nor suggested the claimed invention.

With respect to **Reich,** the reference is directed to plastic articles that can be disposed of merely by outdoor exposure to sunlight so that they do not require incineration, burial, or dumping for disposal. **Reich** primarily teaches materials and methods in which the polypropylene can be made to interact better with the sunlight to achieve certain results. **Reich** teaches that polymers degrade in sunlight and discloses different ways one can stabilize polymers in sunlight. **Reich** further teaches how to make a film for agricultural

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stabilize polymers in sunlight. **Reich** further teaches how to make a film for agricultural purposes that degrades in sunlight using modified polymers. The polymer (polypropylene) in **Reich** is so unstable to the sunlight that one is required to incorporate the disclosed chemicals in the recited polymer to stabilize it in predictable and reliable way so that when one returns to plant the next year, the film is degraded and one does not have to manually going into the ground to remove the polymer film.

Further, Applicants would argue that one ordinarily skilled in the art would not be motivated to combine the teachings of **Andersson et al.** (liquid container) with **Reich** (plastic article exposed to sun light) will to arrive at the present invention as claimed. Quite simply, there is little to no need to provide a liquid container with a polymer coating that degrades at a predictable or stable rate when exposed to solar or other types of radiation. Applicants would argue that biodegradability or compostability are more desirable characteristics for a liquid container. Conversely, biodegradability would not be desirable to use in the material of **Reich**, at least according to the desired characteristics set out in the reference. Applicant would further argue that **Reich**, in fact, teaches away from the instant invention, namely one would not use materials that accelerate or even moderate the rate of degradation of the polymer of the instant invention. Finally, irrespective of what **Reich** teaches with respect to film degradation, the failure of **Andersson** as a primary reference negates the combination of **Andersson** in view of **Reich** as a basis for rejection of claims 74-76, 79 and 84-86 of the present invention. Applicants respectfully request withdrawal of this rejection.

3. Claims 77-78 and 80-83 are rejected under 35 U.S.C. § 103 (a) as being unpatentable over Andersson et al (U.S. Patent Publication No. 2005/0008800 A1) in

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view of Reich et al (USP #3,984,940) and further in view of Gen (USP 6,949, 254).

Andersson in view of **Reich** is discussed hereinabove and thus will not be repeated to avoid redundancy. With respect to **Gen**, this reference is related to the treatment of biodegradable polymer composition that has sufficiently mild properties so that it is suitable for medical treatment of parts of the body and their environment, while maintaining its shape and properties for a necessary period. It should be obvious to one even slightly skilled in the art, that the teaches of **Gen** are not remotely related to the present invention as claimed and thus not applicable to making the rejection. Nonetheless, claims 77-78 and 80-83 are all dependent directly/indirectly on the claim 74 and therefore inherit each and every element of their parent claim and any intervening claim(s) and thus define a patentable subject matter. Applicants respectfully request withdrawal of this rejection.

It is respectfully submitted that none of the references of record, including those applied in rejecting the claims, whether considered individually or in combination, teaches the claimed invention and it would not be obvious to combine the references to produce the claimed invention.

Concluding remarks:

It is respectfully submitted that while it is possible that the various modifications, or at least some of them, as proposed by the examiner *could* be made to the **Andersson**'s patent by selecting them piecemeal from the various prior art references, all the elements of the Applicants' claims are not found in the combination of the cited references, there is no teaching, suggestion or motivation for making them and the proposed modifications are not obvious in the context of 35 USC 103. Moreover, **Andersson**'s patent in view **Reich** in fact teaches away from a degradable polymer selected from the group consisting of a degradable

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polyolefin, a polyhydroxyalkanoate, polylactic acid, and degradable polyester.

In determining the differences between the prior art and the claims, the question under 35 U.S.C. 103 is not whether the differences themselves would have been obvious, but whether the claimed invention as a whole would have been obvious. Stratoflex, Inc. v. Aeroquip Corp., 713 F.2d 1530, 218 USPQ 871 (Fed. Cir. 1983).

As reiterated by the Supreme Court in KSR International Co. v. Teleflex Inc. (KSR), 550 U.S. ___, 82 USPQ2d 1385 (2007), the framework for the objective analysis for determining obviousness under 35 U.S.C. 103 is stated in Graham v. John Deere Co., 383 U.S. 1, 148 USPQ 459 (1966). Obviousness is a question of law based on underlying factual inquiries. Although the Supreme Court in KSR cautioned against an overly rigid application of the teaching, suggestion, motivation (TSM) rationale, it also recognized that TSM was one of a number of valid rationales that could be used to determine obviousness.

A prior art reference must be considered in its entirety, i.e., as a whole, including portions that would lead away from the claimed invention. W.L. Gore & Associates, Inc. v. Garlock, Inc., 721 F.2d 1540, 220 USPQ 303 (Fed. Cir. 1983), cert. denied, 469 U.S. 851 (1984).

The mere fact that a worker in the art could rearrange the parts of the reference device to meet the terms of the claims is not by itself sufficient to support a finding of obviousness. The prior art must provide a motivation or reason for the worker in the art, without the benefit of appellant's specification, to make the necessary changes in the reference device. Ex parte Chicago Rawhide Mfg. Co., 223 USPQ 351, 353 (Bd. Pat. App. & Inter. 1984).

A statement that modifications of the prior art to meet the claimed invention would have been "'well within the ordinary skill of the art at the time the claimed invention was made'" because the references relied upon teach that all aspects of the claimed invention

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were individually known in the art is not sufficient to establish a prima facie case of obviousness without some objective reason to combine the teachings of the references. Ex parte Levengood, 28 USPQ2d 1300 (Bd. Pat. App. & Inter. 1993).

If proposed modification would render the prior art invention being modified unsatisfactory for its intended purpose, then there is no suggestion or motivation to make the proposed modification. In re Gordon, 733 F.2d 900, 221 USPQ 1125 (Fed. Cir. 1984).

If the proposed modification or combination of the prior art would change the principle of operation of the prior art invention being modified, then the teachings of the references are not sufficient to render the claims prima facie obvious. In re Ratti, 270 F.2d 810, 123 USPQ 349 (CCPA 1959).

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CONCLUSION

Based upon the foregoing, Applicants respectfully submit that claims 74-86 particularly define and patentably distinguish the present invention over the cited references. Accordingly, reconsideration of the rejection and allowance of all claims are earnestly requested. However, should the Examiner have any remaining questions and the attending to of which would expedite such action, the Examiner is invited to contact the undersigned at the telephone number listed below.

No month extension of time is believed to be required. However, the Commissioner is authorized to charge any fees associated with this or any other communication, or credit any over payment, to Deposit Account No. 09-0525.

Respectfully Submitted,

Date: January 28, 2010

By: 

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Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
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Electronic Acknowledgement Receipt	
EFS ID:	6901972
Application Number:	11348150
International Application Number:	
Confirmation Number:	5681
Title of Invention:	Biodegradable paper-based laminate with oxygen and moisture barrier properties and method for making biodegradable paper-based laminate
First Named Inventor/Applicant Name:	Christopher S. Cleveland
Customer Number:	01726
Filer:	Matthew M. Eslami/Michele McRoy
Filer Authorized By:	Matthew M. Eslami
Attorney Docket Number:	FDS-029388-US
Receipt Date:	28-JAN-2010
Filing Date:	06-FEB-2006
Time Stamp:	15:55:35
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		100128_rsptooffaction_fds029388us.pdf	184882 d62bb1cbaea313b96892f89792787a64e0ddc316	yes	11

	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	11
Warnings:			
Information:			
	Total Files Size (in bytes):	184882	

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 (07-06)
Approved for use through 1/31/2007. 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	Filing Date	☐ To be Mailed
11/348,150	02/06/2006	

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	
☐ SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A	N/A			N/A	
☐ EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A	N/A			N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	minus 20 =	*	X $ =		OR	X $ =	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	minus 3 =	*	X $ =			X $ =	
☐ APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $250 ($125 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			TOTAL	

APPLICATION AS AMENDED – PART II

AMENDMENT	01/28/2010	(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 ($)	OTHER THAN SMALL ENTITY ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	* 13	Minus	** 73	= 0	X $ =		OR	X $52=	0
	Independent (37 CFR 1.16(h))	* 1	Minus	***13	= 0	X $ =		OR	X $220=	0
	☐ 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	0

AMENDMENT		(Column 1) CLAIMS REMAINING AFTER AMENDMENT		(Column 2) HIGHEST NUMBER PREVIOUSLY PAID FOR	(Column 3) PRESENT EXTRA	RATE ($)	ADDITIONAL FEE ($)		RATE ($)	ADDITIONAL FEE ($)
	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 number found in the appropriate box in column 1.

Legal Instrument Examiner:
/KAREN VESTAL/

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.

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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
11/348,150	02/06/2006	Christopher S. Cleveland	FDS-029388-US	5681

1726 7590 05/12/2010

INTERNATIONAL PAPER COMPANY
6285 TRI-RIDGE BOULEVARD
LOVELAND, OH 45140

EXAMINER	
PATTERSON, MARC A	
ART UNIT	PAPER NUMBER
1782	

MAIL DATE	DELIVERY MODE
05/12/2010	PAPER

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.

Office Action Summary	Application No.	Applicant(s)	
	11/348,150	CLEVELAND ET AL.	
	Examiner	Art Unit	
	MARC A. PATTERSON	1782	

-- 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 MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, 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 *1/28/10*.

2a)☒ This action is **FINAL**. 2b)☐ This action is non-final.

3)☐ 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

4)☒ Claim(s) *74-86* is/are pending in the application.

4a) Of the above claim(s) _____ is/are withdrawn from consideration.

5)☐ Claim(s) _____ is/are allowed.

6)☒ Claim(s) *74-86* is/are rejected.

7)☐ Claim(s) _____ is/are objected to.

8)☐ Claim(s) _____ are subject to restriction and/or election requirement.

Application Papers

9)☐ The specification is objected to by the Examiner.

10)☐ 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).

11)☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

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).

a)☐ All b)☐ Some * c)☐ None of:

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) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
3) ☐ Information Disclosure Statement(s) (PTO/SB/08) Paper No(s)/Mail Date _____.
4) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date. _____ .
5) ☐ Notice of Informal Patent Application
6) ☐ Other: _____.

DETAILED ACTION

REPEATED REJECTIONS

1. The 35 U.S.C. 103(a) rejection of Claims 74 - 76, 79 and 84 – 86 as being unpatentable over Andersson et al. (U.S. Patent Publication No. 2005/0008800) in view of Reich et al (U.S. Patent No. 3,984,940), of record on page 2 of the previous Action, is repeated.

2. The 35 U.S.C. 103(a) rejection of Claims 77 - 78 and 80 – 83 as being unpatentable over Andersson et al. (U.S. Patent Publication No. 2005/0008800) in view of Reich et al (U.S. Patent No. 3,984,940) and further in view of Gen (U.S. Patent No. 6,949,254 B2), of record on page 2 of the previous Action, is repeated.

ANSWERS TO APPLICANT'S ARGUMENTS

3. Applicant's arguments regarding the 35 U.S.C. 103(a) rejection of Claims 74 - 76, 79 and 84 – 86 as being unpatentable over Andersson et al. (U.S. Patent Publication No. 2005/0008800) in view of Reich et al (U.S. Patent No. 3,984,940) and 35 U.S.C. 103(a) rejection of Claims 77 - 78 and 80 – 83 as being unpatentable over Andersson et al. (U.S. Patent Publication No. 2005/0008800) in view of Reich et al (U.S. Patent No. 3,984,940) and further in view of Gen (U.S. Patent No. 6,949,254 B2), of record in the previous Action, have been carefully considered but have not been found to be persuasive for the reasons set forth below.

Applicant argues, on page 5 of the remarks dated January 28, 2010, that the degradability taught by Reich et al is not the same as the degradability disclosed in the specification, which is biodegradability.

However, the claimed invention is not limited to the definition of the specification, or biodegradability. Furthermore, Applicant does not argue that the only reasonable definition of 'degradable' is biodegradable, instead of the degradability taught by Reich et al.

Applicant also argues, on page 6, that Andersson et al disclose non – degradable polyolefins.

However, Andersson et al does not limit the polyolefin to one that is non – degradable.

Applicant also argues on page 6 that Reich et al teach a film and the use of the film for agriculture.

However, Reich et al also teach the film for making a container.

Applicant also argues on page 6 that there is little need to provide a liquid container with a coating that degrades in sunlight, and that biodegradability is more desirable.

However, Applicant does not state why degradation in sunlight is not needed, contrary to the teaching of Reich et al of using the polymer in containers.

4. Applicant's amendment necessitated the new ground(s) of rejection presented in this Office action. Accordingly, **THIS ACTION IS MADE FINAL**. See MPEP § 706.07(a). 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 date of this final action.

5. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Marc A Patterson whose telephone number is 571-272-1497. The examiner can normally be reached on Mon - Fri 8:30 AM - 5:00 PM.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Rena Dye can be reached on 571-272-1498. 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 http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free).

/Marc A Patterson/
Primary Examiner, Art Unit 1782

Docket No.: FDS-029388-US

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF:

CLEVELAND, et al.	:	GROUP ART UNIT: 1782
SERIAL NO: 11/348,150	:	EXAMINER: PATTERSON, MARC A
FILED: February 06, 2006	:	CONFIRMATION #: 5681

FOR: BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOISTURE BARRIER PROPERTIES AND METHOD FOR MAKING BIODEGRADABLE PAPER-BASED LAMINATE

AMENDMENT AFTER FINAL REJECTION PURSUANT TO 37 CFR § 1.116(b)

United States Patent and Trademark Office
Customer Service Window, Mail Stop AF
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

In response to the Office Action (made FINAL) mailed **May 12, 2010**, the period for response which is set to expire August 12, 2010, please amend the above-captioned application, without prejudice or disclaimer as follows, as well as consider the following remarks. Applicants submit there are good and sufficient reasons, in accordance with 37 CFR § 1.116(b), for entering these amendments. In particular, it is believed these amendments will put the above application in better condition for allowance or appeal.

Applicants respectfully request prompt issuance of a Notice of Allowance or an Advisory Action by the Examiner, as the present amendment is being timely filed on or before two months of the mailing date of the Final Action (*i.e.*, on or before July 12, 2009). See MPEP § 706.07(f). Applicants' representative, Matt M. Eslami, also respectfully requests a telephone conference in the event the Examiner has any questions, comments or suggestions that would place any pending claim not in allowable form in better condition for allowance. Applicant's representative may be reached directly at 513-248-6193.

Amendments to the Specification begin on page **2** of this paper.

Amendments to the Claims begin on page **3** of this paper.

Remarks/Arguments begin on page **6** of this paper.

IN THE SPECIFICATION

Please replace paragraph [0014] at page 4, with the following rewritten paragraph:

With the objects of the invention in view, there is also provided a method for making biodegradable paper products with barrier properties. The method includes providing a substrate having a surface and extruding a barrier layer onto the surface of the substrate. The barrier layer includes a degradable material such as polyvinyl alcohol, polylactic acid or polyhydroxyalkanoate ~~polyhydroxyalkonoate~~. The coatweight of the degradable material is within a range from 8 to 20 pounds per three-thousand square feet. A second barrier layer can be extruded on an opposing side of the substrate.

IN THE CLAIMS

The listing of claims will replace all prior versions, and listings, of claims in the application:

Listing of Claims:

Claims 1-73. (Canceled).

74. (Currently Amended) A laminate for forming moisture barriers, comprising:

a substrate having an interior surface for contacting products; and

a moisture protectant layer disposed on said interior surface of said substrate and comprising at least one ~~degradable polymer selected from the group consisting of a degradable polyolefin, a polyhydroxyalkanoate, polylactic acid, and a~~ biodegradable ~~degradable~~ polyester **polymer**.

75. (Currently Amended) The laminate according to claim 74, wherein the moisture protectant layer has a coat weight ~~thickness~~ of from 5 to 30 lbs/3000 ft^2.

76. (Currently Amended) The laminate according to Claim 74, further comprising a second moisture protectant layer disposed on an exterior surface of said substrate and comprising at least one ~~degradable polymer selected from the group consisting of a degradable polyolefin, a polyhydroxyalkanoate, polylactic acid, and a~~ biodegradable ~~degradable~~ polyester **polymer**.

77. (Currently Amended) The laminate according to Claim 88 ~~76~~, wherein the second moisture protectant layer further comprises at least one filler.

78. (Currently Amended) The laminate according to Claim 77 ~~76~~, wherein the second moisture protectant layer ~~further~~ comprises at least one filler ~~member~~ selected from the group consisting of calcium carbonate, starch, clay kaolin, talc, cellulose fibers, and diatomaceous earth.

79. (Currently Amended) The laminate according to claim 76, wherein the second moisture

protectant layer has a coat weight ~~thickness~~ of from 5 to 30 lbs/3000 ft^2.

80. (Previously Presented) The laminate according to Claim 76, wherein the second moisture protectant layer further comprises at least one member selected from the group consisting of calcium carbonate and starch.

81. (Currently Amended) The laminate according to Claim 87 ~~74~~, wherein the moisture protectant layer further comprises at least one filler.

82. (Currently Amended) The laminate according to 81 ~~74~~, wherein the moisture protectant layer ~~further~~ comprises at least one filler ~~member~~ selected from the group consisting of calcium carbonate, starch, clay kaolin, talc, cellulose fibers, and diatomaceous earth.

83. (Previously Presented) The laminate according to Claim 74, wherein the moisture protectant layer further comprises at least one member selected from the group consisting of calcium carbonate and starch.

84. (Currently Amended) The laminate according to Claim 74, wherein the substrate comprises paperboard; and the moisture protectant layer has a coat weight ~~thickness~~ of from 5 to 30 lbs/3000 ft^2 and comprises polylactic acid and at least one member selected from the group consisting of calcium carbonate and starch.

85. (Previously Presented) A cup, comprising the laminate according to Claim 84.

86. (Previously presented) A cup, comprising the laminate according to Claim 74.

87. (New) The laminate according to claim 74, wherein the at least one biodegradable polyester polymer is selected from the group consisting of compostable polylactic acid and a compostable polyhydroxyalkanoate.

88. (New) The laminate according to claim 76, wherein the second moisture protectant layer comprises at least one biodegradable polyester polymer selected from the group consisting of compostable polylactic acid and a compostable polyhydroxyalkanoate.

REMARKS/ARGUMENTS

A. Status of Claims

Favorable reconsideration of this application, as presently amended, is respectfully requested. Claims 1-73 have previously been cancelled. New Claims 87-88 have been added. Claims 74-88 are currently pending.

B. Amendments to Specification and Claims

A typographical error in paragraph [0014] at page 4 of the above application has been corrected.

Claim 74 has been amended to recite that the moisture protectant layer disposed on the interior surface of the substrate comprises at least one **biodegradable**-polyester **polymer**. Support for these amendments to Claim 74 may be found in, for example, paragraphs [0030], [0032]-[0033], [0035], and [0040] of the above application.

Claim 76 has been similarly amended to recite that the second moisture protectant layer disposed on the exterior surface of the substrate comprises at least one **biodegradable** polyester **polymer**. Support for these amendments to Claim 76 may also be found in, for example, paragraphs [0030], [0032]-[0033], [0035], and [0040] of the above application.

Claims 75, 79, and 84 have been amended to change "thickness" to "coat weight." Support for this amendment to Claims 75, 79, and 84 may also be found in, for example, paragraph [0030] of the above application.

New Claims 87 and 88 have been added. New Claim 87 recites that the moisture protectant layer comprises at least one biodegradable polyester polymer selected from the group consisting of **compostable polylactic acid** and **a compostable polyhydroxyalkanoate**, while new Claim 88 similarly recites that the second moisture protectant layer comprises at least one biodegradable polyester polymer selected from the group consisting of **compostable polylactic acid** and a **compostable polyhydroxyalkanoate**. Support for new Claims 87 and 88 may be found in, for example, paragraphs [0009], [0012]-[0013], [0033], [0035], and [0040] of the above application.

Claim 77 has been amended to depend from new Claim 88, while Claim 78 has been amended to depend from amended Claim 77, as well as to recite that the second moisture protectant layer comprises at least one **filler** selected from the group consisting of calcium

carbonate, starch, clay kaolin, talc, cellulose fibers, and diatomaceous earth. Support for this amendment to Claim 78 may be found in, for example, paragraph [0051] of the above application.

Similar to Claim 77, Claim 81 has been amended to depend from new Claim 87, while Claim 82 (similar to amended Claim 78) has been amended to depend from amended Claim 81, as well as to recite that the moisture protectant layer comprises at least one **filler** selected from the group consisting of calcium carbonate, starch, clay kaolin, talc, cellulose fibers, and diatomaceous earth. Support for this amendment to Claim 82 may be found in, for example, paragraph [0051] of the above application.

C. Response to Rejection of Claims under 35 U.S.C. § 103(a) as Being Unpatentable over Andersson, in view of Reich

At pages 2-3 of the Final Office Action ("Final Action"), Claims 74-76, 79, and 84-86 have been rejected under 35 U.S.C. § 103(a) as being unpatentable over U.S. Pat. Appln. No. 2005/0008800 to Andersson et al (hereafter referred to as "**Andersson**"), in view of U.S. Pat. No. 3,984,940 to Reich et al (hereafter referred to as "**Reich**"), referring also to page 2 of the previous October 28, 2009 Office Action (hereafter referred to as "October 28, 2009 Office Action"). **This rejection is respectfully traversed with respect to these Claims**, as amended or as currently presented, for at least the following reasons.

In support of the rejection of Claims 74-76, 79, and 84-86 under 35 U.S.C. § 103(a) as being unpatentable over **Andersson**, in view of **Reich**, the Final Action makes the following allegations:

> ***Allegation 1***: Applicant argues, on page 5 of the remarks dated January 28, 2010 that the degradability taught by **[Reich]** is not the same as the degradability disclosed in the specification, which is **biodegradability**.[1]
>
> ***Allegation 2***: However, **the claimed invention is not limited to the definition of the specification, or biodegradability**. Furthermore, Applicant does not argue that the only reasonable definition of "degradable' is biodegradable, instead of the degradability taught by **[Reich]**.[2]

[1] See page 2 of the Final Action (emphasis added).
[2] See page 2 of the Final Action (emphasis added).

Responsive to Allegations 1 and 2, Claim 74 has been amended to recite that the **moisture protectant layer disposed on the interior surface of the substrate** comprises at least one **biodegradable**-polyester **polymer**. Similarly, Claim 76 (which depends from amended Claim 74) has been amended to recite that the **second moisture protectant layer disposed on the exterior surface of the substrate** comprises at least one **biodegradable**-polyester **polymer**. In other words, each of these respective moisture protectant layers of amended Claims 74 and 76 comprise a **biodegradable** (polyester) polymer. By contrast, **Reich** only teaches polymers of polypropylene **which are degraded by sunlight**,[3] i.e., **are not biodegradable (polyester) polymers** according to amended Claims 74 and 76.

The October 28, 2009 Office Action also admits that "**[Andersson]** fail[s] to disclose a polypropylene that is degradable."[4] In fact, **Andersson fails to teach that any of the polymers disclosed in this reference are "biodegradable"** according to amended Claims 74 and 76. Accordingly, Allegations 1 and 2 no longer establish that **all features** of amended Claims 74 and 76 are taught by the references (i.e., **Andersson** and **Reich**) relied upon in rejecting these Claims. In determining patentability under 35 U.S.C. §103(a) over the art (*i.e.*, the references relied upon in Allegations 1 and 2), **all of the claim limitations** (*e.g.*, claim elements, features, *etc.*) must be considered.[5] **All of the features of** amended Claims 74 and 76 **are not taught or suggested** by **Andersson** and **Reich** because: (a) **Andersson** does not teach any of the polymers disclosed in this reference are **biodegradable**; and (b) **Reich** only teaches polypropylene polymers **which are degraded by sunlight**, i.e., **are not biodegradable polymers** according to amended Claims 74 and 76. Put differently, this rejection of amended Claim 74, as well as amended Claim 76, under 35 U.S.C. § 103(a) over **Andersson**, in view of **Reich**, now fails to satisfy at least one of the criteria for obviousness, namely that the prior art references **must teach or suggest all the claim limitations (e.g., elements, features, etc.)**.

Claims 75 and 84-86, as amended or as currently presented, depend (directly or indirectly) from amended Claim 74. Accordingly, Claims 75 and 84-86 are patentable over **Andersson**, in view of **Reich**, for at least the same reasons amended Claim 74 is patentable

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[3] See abstract of Reich. See also page 2 of October 28, 2009 Office Action which alleges the following: "**[Reich]** teach[es] a polypropylene (column 11, lines 54-61) that is degradable, for the purpose of obtaining a film **that can be degraded by solar radiation** (column 12, lines 10-14) (emphasis added)."

[4] See page 2 of October 28, 2009 Office Action.

[5] See *In re Lowry*, 32 USPQ2d 1031, 1034 (Fed. Cir. 1994).

over these references. Similarly, Claims 79, as amended, depends from amended Claim 76. Accordingly, Claim 79 is patentable over **Andersson**, in view of **Reich**, for at least the same reasons amended Claim 76 is patentable over these references.

In further support of the rejection of Claims 74-76, 79, and 84-86 under 35 U.S.C. § 103(a) as being unpatentable over **Andersson**, in view of **Reich**, the Final Action makes the following additional allegations:

> *Allegation 3*: Applicant argues, on page 6 [of the remarks dated January 28, 2010] that **there is little need to provide a liquid container with a coating that degrades in the sunlight, and that biodegradability is more desirable**.[6]
>
> *Allegation 4*: However, **Applicant does not state why degradation in sunlight is not needed**, contrary to the teaching of [**Reich**] of using the [polypropylene] polymer in containers.[7]

What Allegations 3 and 4 assert also do not support the rejection of Claims 74-76, 79, and 84-86, as amended or as currently presented, as being unpatentable over **Andersson**, in view of **Reich**, for at least the following reasons The asserted basis in the October 28, 2010 Office Action for combining **Reich** with **Andersson** is that "[i]t would have been obvious for one of ordinary skill in the art at the time Applicant's invention was made to have provided for a degradable polypropylene in order **to provide a film that degrades by solar radiation** as taught by [**Andersson**]."[8] But this assertion in the October 28, 2010 Office Action amounts to an **impermissible** "conclusory statement" without any "rational underpinning" as to why one of ordinary skill in the art would combine the teachings of **Reich** with those of **Andersson**, especially in view of what is now defined in amended Claim 74. "Rejections on obviousness grounds cannot be sustained by mere **conclusory statements**; instead there must be some **articulated reasoning** with some **rational underpinning** to support the legal conclusion of obviousness."[9]

---

[6] See page 3 of the Final Action (emphasis added).
[7] See page 3 of the Final Action (emphasis added).
[8] See pages 2-3 of October 28, 2009 Office Action (emphasis added).
[9] *See e.g., In re Kahn*, 78 U.S.P.Q.2d at 1336 (emphasis added), cited with approval in the Supreme Court's decision in *KSR International Co. v. Teleflex, Inc.*, 550 U.S. 398, 418 (2007). In *KSR International*, the Supreme Court also observed "in formulating a rejection under 35 U.S.C. § 103(a) based upon a combination of prior art elements, it remains necessary to identify the reason why a person of ordinary skill in the art would have combined the prior art elements in the manner claimed." *Id.* See also *In re Vaidyanathan* (Fed. Cir.

As asserted in the October 28, 2010 Office Action,[10] Allegations 3 and 4 rely upon the sunlight-degradable polypropylene of **Reich** to allegedly replace polypropylene layer 32 of **Andersson**.[11] But Allegations 3 and 4 **impermissibly ignore** the recitation in amended Claim 74 that the moisture protectant layer comprising the **biodegradable polyester polymer** is "disposed on [the] **interior surface of [the] substrate**" for contacting products. If **Reich** is relied upon by Allegations 3 and 4 to teach a "[polypropylene] film that degrades by solar radiation," there would be no "motivation" for a person of ordinary skill in the art to replace such a film on the **interior surface** of a substrate which is not going to be exposed (or is at least unlikely to be exposed) to sunlight. In other words, Allegations 3 and 4 provide **no factually supported** "rational underpinning" for replacing the polypropylene layer 32 of **Andersson** with the **sunlight-degradable polypropylene film** of **Reich**,[12] much less to suggest that, **based on the teachings of these references**, one of ordinary skill in the art would be "motivated" to use a biodegradable (polyester) polymer in a moisture protectant layer "disposed on [the] **interior surface of [the] substrate**" according to amended Claim 74 (from which Claims 75-76, 79, and 84-86 also ultimately depend).

For at least the foregoing reasons, Claims 74-76, 79, and 84-86, as amended or as currently presented, are patentable over **Andersson**, even in view of **Reich**. Accordingly, the rejection of these Claims under 35 U.S.C. § 103(a) over this combination of references should now be withdrawn.

D. Response to Rejection of Claims 77-78 and 80-83 under 35 U.S.C. § 103(a) as Being Unpatentable over Andersson, in view of Reich

At pages 2-3 of the Final Office Action ("Final Action"), Claims 77-78 and 80-83 have been rejected under 35 U.S.C. § 103(a) as being unpatentable over **Andersson**, in view of **Reich**, and further in view of U.S. Pat. No. 6,949,254 to Gen (hereafter referred to as

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2010)(non-precedential)(slip opinion at 17): "Obviousness is determined as a matter of foresight, not hindsight. [Citations to *KSR International* and *Graham v. John Deere Co.* omitted]. *KSR [International]* did not free the PTO's examination process from explaining its reasoning. In making an obviousness rejection, **the examiner should not rely on conclusory statements that a particular feature of the invention would have been obvious or was well known**. Instead, **the examiner should** elaborate, **discussing the evidence or reasoning that leads the examiner to such a conclusion** (emphasis added).

[10] See pages 2-3 of October 28, 2009 Office Action (emphasis added).

[11] See FIG. 3, as well as paragraphs [0031] and [0036], of **Andersson**.

[12] Also, and unlike the biodegradable **polyester** polymers of amended Claims 74 and 76, the polypropylene of **Andersson** and **Reich** are **polyolefins**.

"**Gen**"), referring also to page 2 of the October 28, 2009 Office Action. **This rejection is respectfully traversed with respect to these**, as amended or as currently presented, for at least the following reasons.

In support of the rejection of Claims 77-78 and 80-83 under 35 U.S.C. § 103(a) as being unpatentable over **Andersson**, in view of **Reich**, and further in view of **Gen**, the Final Action relies upon the following allegations made in the October 28, 2009 Office Action:

> *Allegation 5*: **[Andersson]** and **[Reich]** disclose a laminate as discussed above. The laminate comprises polyester (paragraph 0024). With regard to Claims 77-78 and 80-83, **[Andersson]** and **[Reich]** **fail to disclose polylactic acid and a filler comprising starch**.[13]
>
> *Allegation 6*: **[Gen]** teaches a polyester comprising polylactic acid (column 1, lines 30-31) and starch (column 1, lines 30-31) for the **purpose of using a polymer having good resistance to thermal decomposition**. One of ordinary skill in the art would therefore **have recognized the advantage of providing for the polylactic acid and starch** of **[Gen]** in **[Andersson]** and **[Reich]**, which comprises a polyester, **depending on the desired resistance to thermal decomposition of the end product**.[14]
>
> *Allegation 7*: It therefore would have been obvious for one of ordinary skill in the art at the time Applicant's invention was made to have provided for polylactic acid and starch in **[Andersson]** and **[Reich]** **in order to obtain good resistance to thermal decomposition** as taught by [Gen].[15]

What Allegations 5-7 assert do not support the rejection of Claims 77-78 and 80-83, as amended or as currently presented, as being unpatentable over **Andersson**, in view of **Reich**, and further in view of **Gen**, for at least the following reasons. First, for at least the reasons presented above in response to Allegations 1-4, amended Claims 74 and 76 are patentable over **Andersson**, in view of **Reich**. Allegations 5-7 rely upon **Gen** only to teach the use of starch and polylactic acid to replace the alleged polyester in the alleged laminate of **Andersson** and **Reich**, but not to supply those features (i.e., **biodegradable polyester polymer** which is **disposed on the interior surface** of the substrate) **which are missing from the teachings** of **Andersson** and **Reich**. Claims 77-78 and 80-83 also depend (directly or indirectly) from amended Claims 74 or 76, and are therefore patentable over **Andersson**,

[13] See page 3 of the October 28, 2009 Office Action (emphasis added).
[14] See pages 3-4 of the October 28, 2009 Office Action (emphasis added).
[15] See page 4 of the October 28, 2009 Office Action (emphasis added).

in view of **Reich**, even further in view of **Gen**, for at least the same reasons amended Claims 74 and 76 are patentable over this combination of references.

Second, Claims 77-78 and 80-83 now also depend (directly or indirectly) from new Claims 87 and 88. New Claim 87 depends from amended Claim 74, while new Claim 88 depends from amended Claim 76. Accordingly, new Claims 87 and 88 are patentable over **Andersson**, in view of **Reich**, even further in view of **Gen**, for at least the same reasons amended Claims 74 and 76 are patentable over this combination of references.

Third, new Claim 87 recites that the moisture protectant layer comprises at least one biodegradable polyester polymer selected from **compostable polylactic acid** or a **compostable polyhydroxyalkanoate**, while new Claim 88 similarly recites that the second moisture protectant layer comprises at least one biodegradable polymer selected from **compostable polylactic acid** or a **compostable polyhydroxyalkanoate**. Nowhere do Allegations 5-7 assert that **Andersson**, **Reich**, and/or **Gen** teach: (1) a **compostable polyhydroxyalkanoate** according to new Claims 87 or 88; or (2) at least with regard to **Andersson** and **Reich**, that polylactic acid is **compostable** according to new Claims 87 or 88. In fact, as conceded by Allegation 5, **Andersson** and **Reich** fail to teach polylactic acid, or any other biodegradable or compostable polymer according to new Claims 87 and 88.

As pointed out above in response to Allegations 3 and 4, the Final Action **impermissibly** relies upon the sunlight-degradable polypropylene of **Reich** to allegedly replace polypropylene layer 32 of **Andersson** to assert that it would be obvious to dispose a **biodegradable (polyester) polymer** on the **interior surface of the substrate** according to amended Claim 74. In addition, by suggesting that the alleged polyester of the alleged laminate of **Andersson** and **Reich** can be replaced with the **combination of polylactic acid and starch** (as a filler) allegedly taught by **Gen**, Allegations 5-7 (especially Allegations 6-7) also assert an **impermissible** "conclusory statement" without any "rational underpinning" which is **factually unsupported** by the references relied up with respect to new Claims 87 and 88. In particular, that **Gen** may allegedly teach "a polymer having good resistance to thermal decomposition" is **irrelevant** with respect to disposing on the **interior surface of substrate** a **biodegradable polyester polymer** which is **compostable** polylactic acid (or a **compostable polyhydroxyalkanoate**) according to new Claims 87 and 88. For at least the same reasons, this combination of references asserted by Allegations 5-7 is also

impermissible with respect to Claims 77-78 and 80-83 which depend (directly or indirectly) from new Claims 87 or 88.

Fourth, what Allegations 5-7 assert, as well as suggest, **mischaracterize** what **Gen** fairly teaches with respect to Claims 77-78 and 80-83. Implicit in what Allegations 5-7 assert is that **Gen** teaches the **combination** of polylactic acid and starch (as a filler) according to Claims 77-78 and 80-83. But contrary to what Allegations 5-7 impliedly assert, **Gen does not teach combining polylactic acid with starch as a filler** according to Claims 77-78 and 80-83. The portions of **Gen** relied upon in Allegation 5 for the teachings of polylactic acid and starch do not suggest that these two materials may be combined, and in fact **come from different, unrelated sections in this reference**. In particular, the portion of **Gen** relied upon for teaching starch comes from the Background section of this reference, while the portion of **Gen** relied upon for teaching polylactic acid comes from the Detailed Description of the Invention section. Accordingly, Allegations 5-7 provide **no factually supported basis** in **Gen** for combining polylactic acid with starch (as a filler) according to Claims 77-78 and 80-83.

If the Examiner still maintains that Claims 77-78 and 80-83, as amended or as currently presented, are unpatentable over **Andersson**, in view of **Reich**, and further in view of **Gen**, for the reasons presented in Allegations 5-7, then Applicants assert that this rejection under 35 U.S.C. § 103(a) appears to be based, at least partially, on facts within the Examiner's personal knowledge **which have not been provided to Applicants**. Accordingly, if the Examiner wishes to persist in this rejection of Claims 77-78 and 80-83, as amended or as currently presented, under 35 U.S.C. § 103(a) as unpatentable over **Andersson**, in view of **Reich**, and further in view of **Gen**, for the reasons presented in Allegations 5-7, Applicants **respectfully request** that he provide an affidavit/declaration under 37 CFR § 1.104(d)(2)[16] because this rejection appears to be based, at least partially, on

[16] See 37 CFR § 1.104(d)(2) which states: "When a rejection in an application is based on facts within the personal knowledge of an employee of the Office, the data shall be as specific as possible, and the reference must be supported, when called for by the applicant, by the affidavit of such employee, and such affidavit shall be subject to contradiction or explanation by the affidavits of the applicant and other persons." Applicants note that in the unpublished case of *In re Sun*, 31 USPQ2d 1451, 1455 (Fed. Cir. 1993), the USPTO argued "the procedures established by 37 C.F.R. Section 1.107(b) (1993) [now 37 CFR § 1.104(d)(2)] ***expressly entitle*** an Applicant, on mere request, to an examiner affidavit that provides [citations that support the Examiner's asserted level of skill in the art]" (emphasis added). Furthermore, in *In re Sun*, the Federal Circuit, held that "this procedure, so readily available, helps save the lack of citation in an office action from possible constitutional infirmity in denying reasonable notice and hence due process." See 31 USPQ2d at 1455. See

facts within the Examiner's personal knowledge which have not been provided to Applicants, and which are not fairly taught or suggested by **Andersson**, **Reich**, or **Gen**, or any other verifiable evidence provided by the Examiner.

For at least the foregoing reasons, Claims 77-78 and 80-83, as amended or as currently presented, are patentable over **Andersson**, in view of **Reich**, and even further in view of **Gen**. Accordingly, the rejection of these Claims under 35 U.S.C. § 103(a) over this combination of references should now be withdrawn.

E. Conclusion

Claims 74-86, as amended, as well as new Claims 87-88, are patentable over the art relied upon in the Final Office Action. Accordingly, Claims 74-88 are in condition for allowance, and favorable action is earnestly solicited thereon.

Should the Examiner feel that there are any issues outstanding after consideration of this response, the Examiner is invited to contact Mr. Matt M. Eslami at 513-248-6193 to expedite prosecution of the above application.

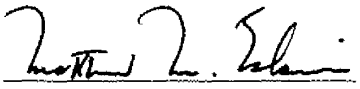
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also *In re Vaidyanathan* (Fed. Cir. 2010)(non-precedential)(slip opinion at 18): "If it is not possible for the examiner to provide this type of information, **the examiner might choose instead to provide an affidavit detailing the examiner's own personal knowledge** (as a person approximating one of ordinary skill in the art) of the technology in question (citing 37 CFR § 1.104(d)(2)) (emphasis added)."

No extension of time is believed to be required. However, the Commissioner is hereby authorized by this paper to charge any fees due under 37 C.F.R. §§ 1.16 and 1.17 which may be required, including any required extension of time fees, or credit any overpayment, to Deposit Account No. 09-0525. **This paragraph is intended to be a CONSTRUCTIVE PETITION FOR EXTENSION OF TIME in accordance with 37 C.F.R. § 1.136(a)(3) if needed.**

Respectfully Submitted,

Date: June 15, 2010

By: 

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Electronic Acknowledgement Receipt	
EFS ID:	7818222
Application Number:	11348150
International Application Number:	
Confirmation Number:	5681
Title of Invention:	Biodegradable paper-based laminate with oxygen and moisture barrier properties and method for making biodegradable paper-based laminate
First Named Inventor/Applicant Name:	Christopher S. Cleveland
Customer Number:	01726
Filer:	Matthew M. Eslami/Michele McRoy
Filer Authorized By:	Matthew M. Eslami
Attorney Docket Number:	FDS-029388-US
Receipt Date:	15-JUN-2010
Filing Date:	06-FEB-2006
Time Stamp:	16:13:37
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		100615_rsptofinaloffact_fds029388us.pdf	361477 c142cd9f33b38dc84ec19812325aff4870829015	yes	15

	Multipart Description/PDF files in .zip description		
	Document Description	**Start**	**End**
	Amendment After Final	1	1
	Specification	2	2
	Claims	3	5
	Applicant Arguments/Remarks Made in an Amendment	6	15
Warnings:			
Information:			
	Total Files Size (in bytes):	361477	

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.

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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.

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PATENT APPLICATION FEE DETERMINATION RECORD

Substitute for Form PTO-875

Application or Docket Number	Filing Date	☐ To be Mailed
11/348,150	02/06/2006	

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	
☐ SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A	N/A			N/A	
☐ EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A	N/A			N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	minus 20 =	*	X $ =		OR	X $ =	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	minus 3 =	*	X $ =			X $ =	
☐ APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $250 ($125 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			TOTAL	

APPLICATION AS AMENDED – PART II

AMENDMENT	06/15/2010	(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 ($)	OTHER THAN SMALL ENTITY ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	* 15	Minus	** 73	= 0	X $ =		OR	X $52=	0
	Independent (37 CFR 1.16(h))	* 1	Minus	***13	= 0	X $ =		OR	X $220=	0
	☐ 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	0

AMENDMENT		(Column 1) CLAIMS REMAINING AFTER AMENDMENT		(Column 2) HIGHEST NUMBER PREVIOUSLY PAID FOR	(Column 3) PRESENT EXTRA	RATE ($)	ADDITIONAL FEE ($)	OR	RATE ($)	ADDITIONAL FEE ($)
	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 number found in the appropriate box in column 1.

Legal Instrument Examiner:
/BRENDA L. TURNER/

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.**

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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
11/348,150	02/06/2006	Christopher S. Cleveland	FDS-029388-US	5681

1726 7590 06/28/2010

INTERNATIONAL PAPER COMPANY
6285 TRI-RIDGE BOULEVARD
LOVELAND, OH 45140

EXAMINER	
PATTERSON, MARC A	
ART UNIT	PAPER NUMBER
1782	

MAIL DATE	DELIVERY MODE
06/28/2010	PAPER

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.

ADVISORY ACTION

Acknowledgement of Applicant's Amendments

1. The amendment made in Claim 74 in the After Final Amendment filed June 15, 2010 has not been entered because the amendment raises a new issue. Claim 74, prior to amendment, was not directed to a polyester that is biodegradable. The application also did not include Claims 87 – 88. The amendment therefore raises issues which would require further search and consideration to be fully considered, and the amendment has therefore not been entered.

ANSWERS TO APPLICANT'S ARGUMENTS

2. Applicant's arguments regarding the 35 U.S.C. 103(a) rejection of Claims 74 - 76, 79 and 84 - 86 as being unpatentable over Andersson et al. (U.S. Patent Publication No. 2005/0008800) in view of Reich et al (U.S. Patent No. 3,984,940) and 35 U.S.C. 103(a) rejection of Claims 77 - 78 and 80 - 83 as being unpatentable over Andersson et al. (U.S. Patent Publication No. 2005/0008800) in view of Reich et al (U.S. Patent No. 3,984,940) and further in view of Gen (U.S. Patent No. 6,949,254 B2), of record in the previous Action, have been carefully considered but have not been found to be persuasive for the reasons set forth below.

Applicant argues, on page 8 of the remarks dated June 15, 2010, that Andersson et al do not disclose a biodegradable polyester.

However, as stated above, Claim 74, prior to amendment, was not directed to a polyester that is biodegradable. The amendment therefore raises issues which would require further search and consideration to be fully considered, and the amendment has therefore not been entered.

Applicant also argues, on page 12, that Gen does not teach the combination of polylactic acid with starch as a filler, because polylactic acid and starch are taught in different sections of the reference.

However, both polylactic acid and starch are taught by Gen, although in different parts of the reference. It therefore would have been obvious for one of ordinary skill in the art to have provided for a polymer having good resistance to thermal decomposition.

3. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Marc A Patterson whose telephone number is 571-272-1497. The examiner can normally be reached on Mon - Fri 8:30 AM - 5:00 PM.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Rena Dye can be reached on 571-272-1498. 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 http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free).

/Marc A Patterson/
Primary Examiner, Art Unit 1782

Advisory Action Before the Filing of an Appeal Brief	**Application No.** 11/348,150	**Applicant(s)** CLEVELAND ET AL.	
	Examiner MARC A. PATTERSON	**Art Unit** 1782	

--The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

THE REPLY FILED 15 June 2010 FAILS TO PLACE THIS APPLICATION IN CONDITION FOR ALLOWANCE.

1. ☒ The reply was filed after a final rejection, but prior to or on the same day as filing a Notice of Appeal. 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. 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.
   Examiner Note: If box 1 is checked, check either box (a) or (b). ONLY CHECK BOX (b) WHEN THE FIRST REPLY WAS FILED WITHIN TWO MONTHS OF THE FINAL REJECTION. 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) 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 37 CFR 41.37(a).

AMENDMENTS

3. ☒ The proposed amendment(s) 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 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.
   The status of the claim(s) is (or will be) as follows:
   Claim(s) allowed: *none*.
   Claim(s) objected to: *none*.
   Claim(s) rejected: *74-86*.
   Claim(s) withdrawn from consideration: *none*.

AFFIDAVIT OR OTHER EVIDENCE

8. ☐ The affidavit or other evidence filed after a 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).
9. ☐ The affidavit or other evidence filed after the date of filing a 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 a good and sufficient reasons why it is necessary and was not earlier presented. See 37 CFR 41.33(d)(1).
10. ☐ 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

11. ☐ The request for reconsideration has been considered but does NOT place the application in condition for allowance because: _____.
12. ☐ Note the attached Information *Disclosure Statement*(s). (PTO/SB/08) Paper No(s). _____
13. ☒ Other: See attached.

/Marc A Patterson/
Primary Examiner, Art Unit 1782

Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
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Docket No.: FDS-029388-US

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF:

CLEVELAND, et al.	:	GROUP ART UNIT: 1782
SERIAL NO: 11/348,150	:	EXAMINER: PATTERSON, MARC A
FILED: February 06, 2006	:	CONFIRMATION #: 5681

FOR: BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOISTURE BARRIER PROPERTIES AND METHOD FOR MAKING BIODEGRADABLE PAPER-BASED LAMINATE

AMENDMENT AFTER FINAL REJECTION PURSUANT TO 37 CFR § 1.116(b)

Do not enter

/M.P./

06/23/2010

United States Patent and Trademark Office
Customer Service Window, Mail Stop AF
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

In response to the Office Action (made FINAL) mailed **May 12, 2010**, the period for response which is set to expire August 12, 2010, please amend the above-captioned application, without prejudice or disclaimer as follows, as well as consider the following remarks. Applicants submit there are good and sufficient reasons, in accordance with 37 CFR § 1.116(b), for entering these amendments. In particular, it is believed these amendments will put the above application in better condition for allowance or appeal.

Applicants respectfully request prompt issuance of a Notice of Allowance or an Advisory Action by the Examiner, as the present amendment is being timely filed on or before two months of the mailing date of the Final Action (*i.e.*, on or before July 12, 2009). See MPEP § 706.07(f). Applicants' representative, Matt M. Eslami, also respectfully requests a telephone conference in the event the Examiner has any questions, comments or suggestions that would place any pending claim not in allowable form in better condition for allowance. Applicant's representative may be reached directly at 513-248-6193.

Amendments to the Specification begin on page **2** of this paper.

Amendments to the Claims begin on page **3** of this paper.

Remarks/Arguments begin on page **6** of this paper.

PTO/SB/30EFS (07-09)
Approved for use through 07/31/2012. 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	11/348,150	Filing Date	2006-02-06	Docket Number (if applicable)	FDS-029388-US	Art Unit	1782
First Named Inventor	Christopher S. Cleveland			Examiner Name	Patterson, Marc A.		

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 Applicant submitted an amendment under Rule 116 on 6/15/2010. The Examiner did not enter the amendment due to raising new issues in the amendment that require the Examiner to conduct a new search. The attached RCE should remove the finality of the OA

☒ 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 090525

SIGNATURE OF APPLICANT, ATTORNEY, OR AGENT REQUIRED

☒ Patent Practitioner Signature

☐ Applicant Signature

PTO/SB/30EFS (07-09)
Approved for use through 07/31/2012. 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	/Matthew M. Eslami/	Date (YYYY-MM-DD)	2010-07-16
Name	Matthew M. Eslami	Registration Number	45488

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.

Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
Page 368 of 1179

PTO/SB/08a (01-10)
Approved for use through 07/31/2012. 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	11348150
	Filing Date	2006-02-06
	First Named Inventor	CLEVELAND, CS
	Art Unit	1782
	Examiner Name	Patterson, Marc A.
	Attorney Docket Number	FDS-029388-US

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	5883199		1999-03-16	McCarthy 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
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FOREIGN PATENT DOCUMENTS Remove

Examiner Initial*	Cite No	Foreign Document Number[3]	Country Code[2] ¡	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							☐

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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	11348150
	Filing Date	2006-02-06
	First Named Inventor	CLEVELAND, CS
	Art Unit	1782
	Examiner Name	Patterson, Marc A.
	Attorney Docket Number	FDS-029388-US

	1		☐

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	11348150
	Filing Date	2006-02-06
	First Named Inventor	CLEVELAND, CS
	Art Unit	1782
	Examiner Name	Patterson, Marc A.
	Attorney Docket Number	FDS-029388-US

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.

☐ Fee set forth in 37 CFR 1.17 (p) has been submitted herewith.

☒ None

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	/Matthew M. Eslami/	Date (YYYY-MM-DD)	2010-07-16
Name/Print	Matthew M. Eslami	Registration Number	45488

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.**

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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.

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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).

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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.

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Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
Page 372 of 1179

Electronic Patent Application Fee Transmittal	
Application Number:	11348150
Filing Date:	06-Feb-2006
Title of Invention:	Biodegradable paper-based laminate with oxygen and moisture barrier properties and method for making biodegradable paper-based laminate
First Named Inventor/Applicant Name:	Christopher S. Cleveland
Filer:	Matthew M. Eslami/Michele McRoy
Attorney Docket Number:	FDS-029388-US
Filed as Large Entity	

Utility under 35 USC 111(a) Filing Fees

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($)
Miscellaneous:				
Request for continued examination	1801	1	810	810
		Total in USD ($)		**810**

Electronic Acknowledgement Receipt	
EFS ID:	8035866
Application Number:	11348150
International Application Number:	
Confirmation Number:	5681
Title of Invention:	Biodegradable paper-based laminate with oxygen and moisture barrier properties and method for making biodegradable paper-based laminate
First Named Inventor/Applicant Name:	Christopher S. Cleveland
Customer Number:	01726
Filer:	Matthew M. Eslami/Michele McRoy
Filer Authorized By:	Matthew M. Eslami
Attorney Docket Number:	FDS-029388-US
Receipt Date:	16-JUL-2010
Filing Date:	06-FEB-2006
Time Stamp:	17:32:18
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	Deposit Account
Payment was successfully received in RAM	$810
RAM confirmation Number	3948
Deposit Account	090525
Authorized User	
The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows: Charge any Additional Fees required under 37 C.F.R. Section 1.16 (National application filing, search, and examination fees) Charge any Additional Fees required under 37 C.F.R. Section 1.17 (Patent application and reexamination processing fees)	

Charge any Additional Fees required under 37 C.F.R. Section 1.19 (Document supply fees)

Charge any Additional Fees required under 37 C.F.R. Section 1.20 (Post Issuance fees)

Charge any Additional Fees required under 37 C.F.R. Section 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)	100716_rce1_fds029388us.pdf	39386 6b18d9242bf7ccc7051a035c9380916a71bef619	no	3
Warnings:					
This is not a USPTO supplied RCE SB30 form.					
Information:					
2	Information Disclosure Statement (IDS) Filed (SB/08)	100716_suppids_fds029388us.pdf	612258 ef5f6fa58077f2a4ebeeb742ee1047cef874ee80	no	4
Warnings:					
Information:					
3	Fee Worksheet (PTO-875)	fee-info.pdf	30389 b28de15d91307ee91684490388b928b7637fddd7	no	2
Warnings:					
Information:					
		Total Files Size (in bytes):	682033		

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.

<u>New Applications Under 35 U.S.C. 111</u>
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.

<u>National Stage of an International Application under 35 U.S.C. 371</u>
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.

<u>New International Application Filed with the USPTO as a Receiving Office</u>
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 (07-06)
Approved for use through 1/31/2007. 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 11/348,150	Filing Date 02/06/2006	☐ To be Mailed

APPLICATION AS FILED – PART I

	(Column 1)	(Column 2)	SMALL ENTITY ☐ RATE ($)	SMALL ENTITY ☐ FEE ($)	OR	OTHER THAN SMALL ENTITY RATE ($)	OTHER THAN SMALL ENTITY FEE ($)
FOR	NUMBER FILED	NUMBER EXTRA					
☐ BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A	N/A			N/A	
☐ SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A	N/A			N/A	
☐ EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A	N/A			N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	minus 20 =	*	X $ =		OR	X $ =	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	minus 3 =	*	X $ =			X $ =	
☐ APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $250 ($125 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			TOTAL	

APPLICATION AS AMENDED – PART II

AMENDMENT	07/16/2010	(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 ($)	OTHER THAN SMALL ENTITY ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	* 15	Minus	** 73	= 0	X $ =		OR	X $52=	0
	Independent (37 CFR 1.16(h))	* 1	Minus	***13	= 0	X $ =		OR	X $220=	0
	☐ 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	0

AMENDMENT		(Column 1) CLAIMS REMAINING AFTER AMENDMENT		(Column 2) HIGHEST NUMBER PREVIOUSLY PAID FOR	(Column 3) PRESENT EXTRA	RATE ($)	ADDITIONAL FEE ($)		RATE ($)	ADDITIONAL FEE ($)
	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 number found in the appropriate box in column 1.

Legal Instrument Examiner:
/DIANE FLOYD/

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.

Docket No.: FDS-029388-US

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF:

CLEVELAND, et al.	:	GROUP ART UNIT: 1782
SERIAL NO: 11/348,150	:	EXAMINER: PATTERSON, MARC A
FILED: February 06, 2006	:	CONFIRMATION #: 5681

FOR: BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOISTURE BARRIER PROPERTIES AND METHOD FOR MAKING BIODEGRADABLE PAPER-BASED LAMINATE

AMENDMENT AFTER FINAL REJECTION PURSUANT TO 37 CFR § 1.116(b)

United States Patent and Trademark Office
Customer Service Window, Mail Stop AF
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

In response to the Office Action (made FINAL) mailed **May 12, 2010**, the period for response which is set to expire August 12, 2010, please amend the above-captioned application, without prejudice or disclaimer as follows, as well as consider the following remarks. Applicants submit there are good and sufficient reasons, in accordance with 37 CFR § 1.116(b), for entering these amendments. In particular, it is believed these amendments will put the above application in better condition for allowance or appeal.

Applicants respectfully request prompt issuance of a Notice of Allowance or an Advisory Action by the Examiner, as the present amendment is being timely filed on or before two months of the mailing date of the Final Action (*i.e.*, on or before July 12, 2009). See MPEP § 706.07(f). Applicants' representative, Matt M. Eslami, also respectfully requests a telephone conference in the event the Examiner has any questions, comments or suggestions that would place any pending claim not in allowable form in better condition for allowance. Applicant's representative may be reached directly at 513-248-6193.

Amendments to the Specification begin on page 2 of this paper.

Amendments to the Claims begin on page 3 of this paper.

Remarks/Arguments begin on page 6 of this paper.

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.
11/348,150	02/06/2006	Christopher S. Cleveland	FDS-029388-US	5681

1726 7590 09/15/2010

INTERNATIONAL PAPER COMPANY
6285 TRI-RIDGE BOULEVARD
LOVELAND, OH 45140

EXAMINER	
PATTERSON, MARC A	

ART UNIT	PAPER NUMBER
1782	

MAIL DATE	DELIVERY MODE
09/15/2010	PAPER

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.

Office Action Summary	Application No.	Applicant(s)	
	11/348,150	CLEVELAND ET AL.	
	Examiner	Art Unit	
	MARC A. PATTERSON	1782	

-- 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 MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, 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 *06 July 2010*.

2a)☐ This action is **FINAL**. 2b)☒ This action is non-final.

3)☐ 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

4)☒ Claim(s) *74-88* is/are pending in the application.

4a) Of the above claim(s) _____ is/are withdrawn from consideration.

5)☐ Claim(s) _____ is/are allowed.

6)☒ Claim(s) *74-88* is/are rejected.

7)☐ Claim(s) _____ is/are objected to.

8)☐ Claim(s) _____ are subject to restriction and/or election requirement.

Application Papers

9)☐ The specification is objected to by the Examiner.

10)☐ 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).

11)☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

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).

a)☐ All b)☐ Some * c)☐ None of:

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) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
3) ☒ Information Disclosure Statement(s) (PTO/SB/08) Paper No(s)/Mail Date *7/16/10*.
4) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date. _____ .
5) ☐ Notice of Informal Patent Application
6) ☐ Other: _____.

Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
Page 380 of 1179

DETAILED ACTION

WITHDRAWN REJECTIONS

1. The 35 U.S.C. 103(a) rejection of Claims 74 - 76, 79 and 84 – 86 as being unpatentable over Andersson et al. (U.S. Patent Publication No. 2005/0008800) in view of Reich et al (U.S. Patent No. 3,984,940), of record on page 2 of the previous Action, is withdrawn.

2. The 35 U.S.C. 103(a) rejection of Claims 77 - 78 and 80 – 83 as being unpatentable over Andersson et al. (U.S. Patent Publication No. 2005/0008800) in view of Reich et al (U.S. Patent No. 3,984,940) and further in view of Gen (U.S. Patent No. 6,949,254 B2), of record on page 2 of the previous Action, is withdrawn.

NEW REJECTIONS

Claim Rejections – 35 USC § 102(b)

3. The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

> A person shall be entitled to a patent unless –
>
> (b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of application for patent in the United States.

4. Claims 74, 76 – 78, 80 – 83 and 85 – 88 are rejected under 35 U.S.C. 102(b) as being anticipated by Doane et al. (U.S. Patent No. 6,040,063)

With regard to Claims 74 and 76, Doane et al discloses a laminate comprising substrate comprising a substrate (column 4, lines 32 – 39) and an interior moisture protective layer

(coating '16' in Figure 2; column 4, lines 29 – 31) comprising biodegradable polyester (column 4, line 52)

With regard to Claims 77 – 78, 80 – 83 and 87 – 88, Doane et al discloses polyester comprising polyhydroxyalkanoate (column 10, line 42) and a filler comprising calcium carbonate (column 9, line 41).

With regard to Claims 85 - 86, the laminate is a cup (tray; column 4, lines 10 – 13).

Claim Rejections – 35 USC § 103(a)

5. The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

> (a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negatived by the manner in which the invention was made.

6. Claims 75, 79 and 84 are rejected under 35 U.S.C. 103(a) as being unpatentable over Doane et al. (U.S. Patent No. 6,040,063).

Doane et al disclose a laminate as discussed above. With regard to Claims 75, 79 and 84, Doane et al fail to disclose a polymeric protectant layer, moisture protective layer and product contact layer having a thickness of from five to thirty pounds per three thousand feet. However, Doane et al discloses layers comprised in a tray (column 4, lines 10 - 13), as discussed above. It therefore would have been obvious for one of ordinary skill in the art to have provided for a desired coat weight of polymeric protectant layer, moisture protective layer and product contact layer depending on the desired weight of the package.

ANSWERS TO APPLICANT'S ARGUMENTS

7. Applicant's arguments regarding the 35 U.S.C. 103(a) rejection of Claims 74 - 76, 79 and 84 – 86 as being unpatentable over Andersson et al. (U.S. Patent Publication No. 2005/0008800) in view of Reich et al (U.S. Patent No. 3,984,940) and 35 U.S.C. 103(a) rejection of Claims 77 - 78 and 80 – 83 as being unpatentable over Andersson et al. (U.S. Patent Publication No. 2005/0008800) in view of Reich et al (U.S. Patent No. 3,984,940) and further in view of Gen (U.S. Patent No. 6,949,254 B2), of record in the previous Action, have been considered and have been found to be persuasive. The rejections are therefore withdrawn. The new rejections above are directed to amended Claims 74 - 88.

8. Any inquiry concerning this communication or earlier communications from the examiner should be directed to Marc A Patterson whose telephone number is 571-272-1497. The examiner can normally be reached on Mon - Fri 8:30 AM - 5:00 PM.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Rena Dye can be reached on 571-272-1498. 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 http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free).

/Marc A Patterson/
Primary Examiner, Art Unit 1782

Notice of References Cited	Application/Control No. 11/348,150	Applicant(s)/Patent Under Reexamination CLEVELAND ET AL.	
	Examiner MARC A. PATTERSON	Art Unit 1782	Page 1 of 1

U.S. PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Name	Classification
*	A	US-6,040,063	03-2000	Doane et al.	428/532
	B	US-			
	C	US-			
	D	US-			
	E	US-			
	F	US-			
	G	US-			
	H	US-			
	I	US-			
	J	US-			
	K	US-			
	L	US-			
	M	US-			

FOREIGN PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Country	Name	Classification
	N					
	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.

PTO/SB/08a (01-10)
Approved for use through 07/31/2012. 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	11348150
Filing Date	2006-02-06
First Named Inventor	CLEVELAND, CS
Art Unit	1782
Examiner Name	Patterson, Marc A.
Attorney Docket Number	FDS-029388-US

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
/M.P./	1	5883199		1999-03-16	McCarthy 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
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	1							☐

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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	11348150
	Filing Date	2006-02-06
	First Named Inventor	CLEVELAND, CS
	Art Unit	1782
	Examiner Name	Patterson, Marc A.
	Attorney Docket Number	FDS-029388-US

	1		☐

If you wish to add additional non-patent literature document citation information please click the Add button Add

EXAMINER SIGNATURE

Examiner Signature	/Marc Patterson/	Date Considered	09/10/2010

*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.

UNITED STATES PATENT AND TRADEMARK OFFICE

UNITED STATES DEPARTMENT OF COMMERCE
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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
11/348,150	02/06/2006	Christopher S. Cleveland	FDS-029388-US	5681

1726 7590 12/03/2010

INTERNATIONAL PAPER COMPANY
6285 TRI-RIDGE BOULEVARD
LOVELAND, OH 45140

EXAMINER	
PATTERSON, MARC A	
ART UNIT	PAPER NUMBER
1782	

MAIL DATE	DELIVERY MODE
12/03/2010	PAPER

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.

Interview Summary	Application No.	Applicant(s)	
	11/348,150	CLEVELAND ET AL.	
	Examiner	Art Unit	
	MARC A. PATTERSON	1782	

All participants (applicant, applicant's representative, PTO personnel):

(1) *Marc Patterson*.

(2) *Eric Guttag, Rick Tedford*.

(3)*Tom Ryan, Matt Eslami*.

(4)*Rena Dye*.

Date of Interview: *11/29//10*.

Type: a)☒ Telephonic b)☐ Video Conference
c)☐ Personal [copy given to: 1)☐ applicant 2)☐ applicant's representative]

Exhibit shown or demonstration conducted: d)☐ Yes e)☒ No.
If Yes, brief description: _____.

Claim(s) discussed: *all of the record*.

Identification of prior art discussed: *all of the record*.

Agreement with respect to the claims f)☐ was reached. g)☒ was not reached. h)☐ N/A.

Substance of Interview including description of the general nature of what was agreed to if an agreement was reached, or any other comments: *The claimed invention vs the prior art of record, specifically whether the structural limitations of the claimed invention are disclosed by the prior art of record*.

(A fuller description, if necessary, and a copy of the amendments which the examiner agreed would render the claims allowable, if available, must be attached. Also, where no copy of the amendments that would render the claims allowable is available, a summary thereof must be attached.)

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. See Summary of Record of Interview requirements on reverse side or on attached sheet.

/Marc A Patterson/ Primary Examiner, Art Unit 1782	

Clearwater Paper Corp. - EX1002
Clearwater Paper Corp. v. Graphic Packaging International LLC
Page 389 of 1179

Summary of Record of Interview Requirements

Manual of Patent Examining Procedure (MPEP), Section 713.04, Substance of Interview Must be Made of Record
A complete written statement as to the substance of any face-to-face, video conference, or telephone interview with regard to an application must be made of record in the application whether or not an agreement with the examiner was reached at the interview.

Title 37 Code of Federal Regulations (CFR) § 1.133 Interviews
Paragraph (b)

In every instance where reconsideration is requested in view of an interview with an examiner, a complete written statement of the reasons presented at the interview as warranting favorable action must be filed by the applicant. An interview does not remove the necessity for reply to Office action as specified in §§ 1.111, 1.135. (35 U.S.C. 132)

37 CFR §1.2 Business to be transacted in writing.
All business with the Patent or Trademark Office should be transacted in writing. The personal attendance of applicants or their attorneys or agents at the Patent and Trademark Office is unnecessary. The action of the Patent and Trademark Office will be based exclusively on the written record in the Office. No attention will be paid to any alleged oral promise, stipulation, or understanding in relation to which there is disagreement or doubt.

———

The action of the Patent and Trademark Office cannot be based exclusively on the written record in the Office if that record is itself incomplete through the failure to record the substance of interviews.

It is the responsibility of the applicant or the attorney or agent to make the substance of an interview of record in the application file, unless the examiner indicates he or she will do so. It is the examiner's responsibility to see that such a record is made and to correct material inaccuracies which bear directly on the question of patentability.

Examiners must complete an Interview Summary Form for each interview held where a matter of substance has been discussed during the interview by checking the appropriate boxes and filling in the blanks. Discussions regarding only procedural matters, directed solely to restriction requirements for which interview recordation is otherwise provided for in Section 812.01 of the Manual of Patent Examining Procedure, or pointing out typographical errors or unreadable script in Office actions or the like, are excluded from the interview recordation procedures below. Where the substance of an interview is completely recorded in an Examiners Amendment, no separate Interview Summary Record is required.

The Interview Summary Form shall be given an appropriate Paper No., placed in the right hand portion of the file, and listed on the "Contents" section of the file wrapper. In a personal interview, a duplicate of the Form is given to the applicant (or attorney or agent) at the conclusion of the interview. In the case of a telephone or video-conference interview, the copy is mailed to the applicant's correspondence address either with or prior to the next official communication. If additional correspondence from the examiner is not likely before an allowance or if other circumstances dictate, the Form should be mailed promptly after the interview rather than with the next official communication.

The Form provides for recordation of the following information:

- Application Number (Series Code and Serial Number)
- Name of applicant
- Name of examiner
- Date of interview
- Type of interview (telephonic, video-conference, or personal)
- Name of participant(s) (applicant, attorney or agent, examiner, other PTO personnel, etc.)
- An indication whether or not an exhibit was shown or a demonstration conducted
- An identification of the specific prior art discussed
- An indication whether an agreement was reached and if so, a description of the general nature of the agreement (may be by attachment of a copy of amendments or claims agreed as being allowable). Note: Agreement as to allowability is tentative and does not restrict further action by the examiner to the contrary.
- The signature of the examiner who conducted the interview (if Form is not an attachment to a signed Office action)

It is desirable that the examiner orally remind the applicant of his or her obligation to record the substance of the interview of each case. It should be noted, however, that the Interview Summary Form will not normally be considered a complete and proper recordation of the interview unless it includes, or is supplemented by the applicant or the examiner to include, all of the applicable items required below concerning the substance of the interview.

A complete and proper recordation of the substance of any interview should include at least the following applicable items:

1) A brief description of the nature of any exhibit shown or any demonstration conducted,
2) an identification of the claims discussed,
3) an identification of the specific prior art discussed,
4) an identification of the principal proposed amendments of a substantive nature discussed, unless these are already described on the Interview Summary Form completed by the Examiner,
5) a brief identification of the general thrust of the principal arguments presented to the examiner,
   (The identification of arguments need not be lengthy or elaborate. A verbatim or highly detailed description of the arguments is not required. The identification of the arguments is sufficient if the general nature or thrust of the principal arguments made to the examiner can be understood in the context of the application file. Of course, the applicant may desire to emphasize and fully describe those arguments which he or she feels were or might be persuasive to the examiner.)
6) a general indication of any other pertinent matters discussed, and
7) if appropriate, the general results or outcome of the interview unless already described in the Interview Summary Form completed by the examiner.

Examiners are expected to carefully review the applicant's record of the substance of an interview. If the record is not complete and accurate, the examiner will give the applicant an extendable one month time period to correct the record.

Examiner to Check for Accuracy

If the claims are allowable for other reasons of record, the examiner should send a letter setting forth the examiner's version of the statement attributed to him or her. If the record is complete and accurate, the examiner should place the indication, "Interview Record OK" on the paper recording the substance of the interview along with the date and the examiner's initials.

Docket No.: FDS-029388-US

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF:

CLEVELAND, et al.	:	GROUP ART UNIT: 1794
SERIAL NO: 11/348,150	:	EXAMINER: PATTERSON, MARC A.
FILED: February 06, 2006	:	CONFIRMATION #: 5681

FOR: BIODEGRADABLE PAPER-BASED LAMINATE WITH OXYGEN AND MOISTURE BARRIER PROPERTIES AND METHOD FOR MAKING BIODEGRADABLE PAPER-BASED LAMINATE

AMENDMENT

United States Patent and Trademark Office
Customer Service Window, Mail Stop Amendment
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir:

In response to the Office Action (non-final) mailed September 15, 2010, the period for response which is set to expire December 15, 2010, please amend the above-identified application as follows, as well as consider the following remarks.

Amendments to/Listing of the Claims begin on page **2** of this paper.

Remarks/Arguments begin on page **5** of this paper.

IN THE CLAIMS

The listing of claims will replace all prior versions, and listings, of claims in the application:

Listing of Claims:

Claims 1-73. (Canceled).

74. (Currently Amended) A laminate for forming a moisture barrier [barriers], comprising:

a substrate having ~~an~~ a separate interior product contacting surface [for contacting products] and a separate exterior surface; and

a moisture protectant layer disposed solely on said interior surface of said substrate and comprising at least one biodegradable polyester polymer.

75. (Previously Presented) The laminate according to claim 74, wherein the moisture protectant layer has a coat weight of from 5 to 30 lbs/3000 ft^2.

76. (Currently Amended) [The] A laminate for forming moisture barriers [according to Claim 74], [further] comprising:

a substrate having a separate interior product contacting surface and a separate exterior surface;

a first moisture barrier layer disposed on said interior surface of said substrate; and

a second moisture protectant layer disposed on ~~an~~ said exterior surface of said substrate and comprising at least one biodegradable polyester polymer, wherein the second moisture barrier layer is separate from the first moisture barrier layer disposed on said interior surface of said substrate.

77. (Previously Presented) The laminate according to Claim 88, wherein the second moisture protectant layer further comprises at least one filler.

78. (Previously Presented) The laminate according to Claim 77, wherein the second moisture protectant layer comprises at least one filler selected from the group consisting of

calcium carbonate, starch, clay kaolin, talc, cellulose fibers, and diatomaceous earth.

79. (Previously Presented) The laminate according to claim 76, wherein the second moisture protectant layer has a coat weight of from 5 to 30 lbs/3000 ft^2.

80. (Previously Presented) The laminate according to Claim 76, wherein the second moisture protectant layer further comprises at least one filler selected from the group consisting of calcium carbonate and starch.

81. (Previously Presented) The laminate according to Claim 87, wherein the moisture protectant layer further comprises at least one filler.

82. (Previously Presented) The laminate according to 81, wherein the moisture protectant layer comprises at least one filler selected from the group consisting of calcium carbonate, starch, clay kaolin, talc, cellulose fibers, and diatomaceous earth.

83. (Previously Presented) The laminate according to Claim 74, wherein the moisture protectant layer further comprises at least one member selected from the group consisting of calcium carbonate and starch.

84. (Previously Presented) The laminate according to Claim 74, wherein the substrate comprises paperboard; and the moisture protectant layer has a coat weight of from 5 to 30 lbs/3000 ft^2 and comprises polylactic acid and at least one member selected from the group consisting of calcium carbonate and starch.

85. (Previously Presented) A cup, comprising the laminate according to Claim 84.

86. (Previously presented) A cup, comprising the laminate according to Claim 74.

87. (Previously Presented) The laminate according to claim 74, wherein the at least one biodegradable polyester polymer is selected from the group consisting of compostable polylactic acid and a compostable polyhydroxyalkanoate.

88. (Previously Presented) The laminate according to claim 76, wherein the second moisture protectant layer comprises at least one biodegradable polyester polymer selected from the group consisting of compostable polylactic acid and a compostable polyhydroxyalkanoate.

REMARKS/ARGUMENTS

A. Status of Claims

Favorable reconsideration of this application is respectfully requested. Claims 1-73 have previously been cancelled. Claims 74-88 are currently pending.

B. Procedural Matters

Applicants gratefully acknowledge the Examiner's statements at pages 2 and 4 of the present Office Action that the prior rejection of Claims 74-76, 79, and 84-86 under 35 U.S.C. § 103(a) as being unpatentable over U.S. Pat. Appln. No. 2005/0008800 to Andersson et al (hereafter referred to as "**Andersson**"), in view of U.S. Pat. No. 3,984,940 to Reich et al (hereafter referred to as "**Reich**") has been withdrawn in view of the persuasiveness of Applicants' arguments in their prior amendment of July 6, 2010.

Applicants also gratefully acknowledge the Examiner's statements at pages 2 and 4 of the present Office Action that the prior rejection of Claims 77-78 and 80-83 have been rejected under 35 U.S.C. § 103(a) as being unpatentable over **Andersson**, in view of **Reich**, and further in view of U.S. Pat. No. 6,949,254 to Gen (hereafter referred to as "**Gen**") has been withdrawn in view of the persuasiveness of Applicants' arguments in their prior amendment of July 6, 2010.

Applicants further gratefully acknowledge the courtesy of the telephone interview of November 29, 2010 (hereafter referred to as "November 29, 2010 Interview") extended by Primary Examiner Patterson and Supervisory Primary Examiner (SPE) to Applicants' representatives, Matthew M. Eslami, Thomas W. Ryan, and Eric W. Guttag, where the following issues were discussed: (1) the rejection Claim 74 under 35 U.S.C. § 102(b) as being anticipated by U.S. Pat. No. 6,040,063 to Doane et al. (hereafter "**Doane**"); and (2) the rejection of Claims 77-78 and 80-83 as also under 35 U.S.C. § 102(b) as being anticipated by **Doane**. Applicants also gratefully acknowledge the courtesy of the follow up telephone interview of December 7, 2010 (hereafter referred to as "December 7, 2010 Interview") extended by Primary Examiner Patterson to Applicants' representative, and Eric W. Guttag, where proposed amendments to Claims 74 and 76 were discussed for distinguishing over the **Doane** reference. The substance of these two telephonic interviews is discussed further below, as appropriate.

C. Amendments to Claims

Claim 74 has been amended to recite a laminate for forming **a** moisture **barrier** which comprises a substrate having a **separate** interior **product contacting** surface **and a separate exterior surface**, wherein the moisture protectant layer is disposed **solely** on the interior surface of the substrate. Support for the amendments to Claim 74 may be found in, for example, FIG. 6 and paragraph [0048] of the present application.

Claim 76 has been amended to be in independent form and to recite a laminate **for forming moisture barriers**, comprising: **a substrate having a separate interior product contacting surface and a separate exterior surface; a first moisture barrier layer disposed on said interior surface of said substrate**; and a second moisture protectant layer disposed on said exterior surface of said substrate and comprising at least one biodegradable polyester polymer, **wherein the second moisture barrier layer is separate from the first moisture barrier layer disposed on said interior surface of said substrate**. Support for the amendments to Claim 76 may be found in, for example, FIG. 7 and paragraph [0049] of the present application.

D. Response to Rejection of Claims 74, 76-78, 80-83, and 85-88 under 35 U.S.C. § 102(b) as Being Anticipated by Doane

At pages 2-3 of the present Office Action, Claims 74, 76-78, 80-83, and 85-88 have been rejected under 35 U.S.C. § 102(b) as being anticipated by U.S. Pat. No. 6,040,063 to Doane et al (hereafter referred to as "**Doane**"). **This rejection is respectfully traversed with respect to these Claims**, as currently presented, for at least the following reasons.

In support of the rejection of Claims 74 and 76 as being anticipated by **Doane**, the present Office Action makes the following allegation:

> ***Allegation 1***: [**Doane**] discloses a laminate comprising [a] substrate (column 4, lines 32-39) and **an interior moisture protective layer** (coating '16' in Figure 2; column 4, lines 29-31) comprising [a] biodegradable polyester (column 4, line 52).[1]

Responsive to what Allegation 1 asserts, and especially in view of the how Examiner Patterson has construed what **Doane** teaches at the November 29, 2010 and December 7,

[1] See pages 2-3 of the present Office Action (emphasis added).

2010 Interviews with respect to relationship of layer 16 and structure 12 in tray 10 shown in FIGS. 1 and 2 of **Doane**, Claims 74 and 76 have been amended. Claim 74 has been amended to recite that the substrate has a **separate interior product contacting** surface **and a separate exterior surface**, wherein the moisture protectant layer is disposed **solely** on the interior surface of the substrate. Claim 76 has been amended to recite that the substrate also has a **separate interior product contacting** surface **and a separate exterior surface**; **a first moisture barrier layer disposed on said interior surface of said substrate**; and a second moisture protectant layer disposed on said exterior surface of said substrate, wherein the second moisture barrier layer **is separate from the moisture barrier layer disposed on said interior surface of said substrate**

As construed by Examiner Patterson at the November 29, 2010 Interview, and as asserted by Allegation 1, Examiner Patterson considers coating layer 16 to be the "interior moisture barrier layer" with respect to structure (substrate) 12 and tray 10. But, as was pointed out at the December 7, 2010, coating layer 16, in fact, forms **one continuous and surrounding exterior layer** with respect to structure 12, with structure 12 having **one continuous and exterior surface**. By contrast, Claim 74, as amended, now recites a **separate interior product contacting** surface on which the moisture protectant layer is disposed, as well as a **separate exterior surface**. In addition, coating layer 16 **surrounds** structure 12 of **Doane**, but is not is **disposed on** structure 12. By contrast, the moisture barrier layer is **disposed on** the interior substrate surface in amended Claim 74.

Following up on suggestions made by Applicants' representative that were discussed during the December 7, 2010 Interview, Claim 74 has been further amended to recite that the moisture barrier layer is disposed **solely** on the separate interior product contacting surface. In other words, the moisture barrier layer **does not form one continuous layer that is disposed on both the interior and exterior surface** of the substrate, and is thus unlike **Doane's** coating layer 16 which forms **one continuous and surrounding exterior layer** with respect structure 12. At the December 7, 2010 Interview, Examiner Patterson also agreed that having Claim 74 recite that the moisture barrier layer is disposed **solely** on the separate interior product contacting surface would distinguish Claim 74 over what is taught by **Doane**.

As was discussed at the December 7, 2010 Interview, amended Claim 76 is also

distinguishable over **Doane** in now reciting that the second moisture barrier layer disposed on the exterior substrate surface **is separate from the first moisture barrier layer disposed on the interior substrate surface**. In other words, amended Claim 76 recites **two separate** and **distinct moisture barrier layers**, **one each disposed on the interior and exterior surface of the substrate**. By contrast, coating layer 16 of **Doane** forms only **one continuous and surrounding exterior layer** with respect structure 12.

In support of the rejection of Claims 77-78, 80-83, and 87-88 as being anticipated by **Doane**, the present Office Action makes the following allegation:

> ***Allegation 2***: [**Doane**] discloses [a] polyester comprising polyhydroxyalkanoate (column 10, line 42) and a **filler** comprising calcium carbonate (column 9, line 41).[2]

What Allegation 2 asserts also does not support the rejection of Claims 77-78, 80-83, and 87-88 as being anticipated by **Doane**. Claims 81-83 and 87 depend, directly or indirectly, from amended Claim 74, while Claims 77-78 and 88 depend, directly or indirectly, from amended Claim 76. Accordingly, Claim 77-78, 80-83, and 87-88 are patentable over **Doane** for at least the same reasons presented above in response to Allegation 1, as well as during the December 7, 2010 Interview for why amended Claims 74 and 76 are patentable over this reference.

In addition, Claims 77-78 and 80-83 are further distinguishable over **Doane** in reciting that the moisture protectant layer comprises **at least one filler** which may be calcium carbonate (see Claims 78, 80, and 82). First, Allegation 2 has **mischaracterized** what is fairly taught by **Doane** with regard to the use of calcium carbonate. Contrary to what Allegation 2 asserts, **Doane** does not teach that calcium carbonate is being used as a filler according to Claims 77-78 and 80-83. Instead, **Doane** teaches that calcium carbonate is being used as a "nucleating agent" for a foamable composition.[3]

Second, these "nucleating agents" are taught by **Doane** for use **in preparing natural polymer foams for structure 12**,[4] and not hydroxyfunctional polyester layer 14 which is self-adhered **to the exterior surface** of structure 12. In other words, and contrary to what

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[2] See page 3 of the present Office Action (emphasis added).
[3] See column 9, lines 33-41, of **Doane**.
[4] See, for example, column 7, lines 53-56, and column 9, lines 4-6, of **Doane**.

Allegation 2 asserts, **Doane** does not teach inclusion of calcium carbonate (or any other disclosed "nucleating agent") in its hydroxyfunctional polyester layer 14 (or layer 16), much less the moisture protectant layers of Claims 77-78 and 80-83. In fact, Examiner Patterson agreed at the November 29, 2010 Interview that **Doane** does not teach inclusion of calcium carbonate or any other "filler" in the moisture protectant layers of Claims 77-78 and 80-83.

In support of the rejection of Claims 85-86 as being anticipated by **Doane**, the present Office Action makes the following allegation:

> ***Allegation 3***: The laminate is a **cup** (column 10, line 42).[5]

What Allegation 3 asserts does not support the rejection of Claims 85-86 as being anticipated by **Doane**. Claims 85-86 depend, directly or indirectly, from amended Claim 74. Accordingly, Claim 85-86 are patentable over **Doane** for at least the same reasons presented above in response to Allegation 1, as well as during the December 7, 2010 Interview for why amended Claim 74 is patentable over this reference. In addition, Claims 85-86 are further distinguishable over **Doane** in reciting a **cup** that comprises the laminate of Claim 84 or amended Claim 74. Contrary to what Allegation 3 asserts, one of ordinary skill in the art would understand that the "tray" taught by Doane is not the "cup" recited in Claims 85-86. In fact, **nowhere** does **Doane** teach that its laminates may be used in cups.

For at least the foregoing reasons, Claims 74, 76-78, 80-83, and 85-856, as amended or as currently presented, are patentable over **Doane**. Accordingly, the rejection of these Claims under 35 U.S.C. § 102(b) based on this reference should be withdrawn.

E. Response to Rejection of Claims 75, 79, and 84 under 35 U.S.C. § 103(a) as Being Unpatentable over Doane

At pages 3-4 of the present Office Action, Claims 75, 79, and 84 have been rejected under 35 U.S.C. § 103(a) as being unpatentable over **Doane**. **This rejection is respectfully traversed with respect to these Claims**, as currently presented, for at least the following reasons.

In support of the rejection of Claims 75, 79, and 84 under 35 U.S.C. § 103(a) as being unpatentable over **Doane**, the present Office Action makes the following allegation:

[5] See page 3 of the present Office Action (emphasis added).

> ***Allegation 4***: [**Doane**] discloses a laminate as discussed above. With regard to Claims 75, 79 and 84, [**Doane**] **fails to disclose** a polymeric protectant layer, moisture protective layer and product contact layer having a **thickness** of from five to thirty pounds per three thousand feet. However, [**Doane**] discloses layers comprised in a tray (column 4, lines 10-13), as discussed above. **It therefore would have been obvious** for one of ordinary skill in the art **to have provided for a desired coat weight of polymeric protectant layer, moisture protective layer and product contact layer depending on the desired weight of the package**.[6]

What Allegation 4 asserts does not support the rejection of Claims 75, 79, or 84 as being unpatentable over **Doane**, for at least the following reasons. First, for at least the reasons presented above in response to Allegation 1, amended Claims 74 and 76 are patentable over **Doane**. Allegation 4 does not assert that those features (i.e., in the case of amended Claim 74, a moisture barrier layer **which is disposed only on the interior product contacting surface** of the substrate, and in the case of amended Claim 76, the second moisture barrier layer disposed on the exterior substrate surface **is separate from the firest moisture barrier layer disposed on the interior substrate surface**) which were asserted **to be missing from the teachings** of **Doane** are now taught by that reference. Claims 75, 79, and 84 also depend directly from either amended Claims 74 or 76, and are therefore patentable over **Doane** for at least the same reasons presented above, as well as during the December 7, 2010 Interview for why amended Claims 74 and 76 are patentable over this reference.

Second, Allegation 4 concedes that the coat weights (not **thickness** as asserted by Allegation 4) recited in Claims 75, 79, or 84 are not taught by **Doane**. Instead, Allegation 4 asserts that it "would be obvious . . . to have provided for a desired coat weight of polymeric protectant layer, moisture protective layer and product contact layer depending on the desired weight of the package." That assertion amounts to an **impermissible** "conclusory statement" without any "rational underpinning" which is **factually unsupported** by **Doane**, the reference relied upon in rejecting Claims 75, 79, and 84. "Rejections on obviousness grounds cannot be sustained by mere **conclusory statements**; instead there must be some **articulated reasoning** with some **rational underpinning** to support the legal conclusion of obviousness."[7] Allegation 4 points to **absolutely nothing** in **Doane** (or any other verifiable

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[6] See page 4 of the present Office Action (emphasis added).
[7] *See e.g., In re Kahn,* 78 U.S.P.Q.2d at 1336 (emphasis added), cited with approval in the Supreme Court's decision in *KSR International Co. v. Teleflex, Inc.*, 550 U.S. 398, 418 (2007). In *KSR International*, the