to restore mercury capture capacity. The first of these steps can remove the mercury, but it is unclear whether it actually removes the sulfate poison. The second of these steps is rather expensive, because one is actually reconstituting the reagent on the sorbent

Porous beds containing a mercury oxidizing reagent on a solid support are described in several patents for removal of mercury from gas streams. These include peroxomonosulfate (for example, see U.S. Patent 4,786,483), triiodide or other mixed halogens (for example, see U.S. Patents 3,194,629; 3,662,523), or sulfur (for example, see U.S. Patents 3,194,629, 4,101,631, 4,708,853, 6,258,334). The reagent material is destroyed either by reaction with the flue gas during sorption or by attempts to regenerate the sorbent; there is no way to regenerate these sorbents, except by reimpregnation of the expensive reagent.

Amalgamating noble metals (gold, silver) on a suitable support can be regenerated by microwave heating (for example, see U.S. Patent 6,136,072), but they are expensive and not especially active for sorption in flue gas.

Hg Sorbents for Air or Natural Gas Treatment

Melkersson (U.S Patent 3786619) described a Hg sorbent comprising SeO_2 or SeS_2 for air treatment. This method did not appear to employ a carbon support or halide.

Yan (U.S. Patent 4814152) described a Hg sorbent comprising elemental sulfur and a metal catalyst on a carbon support. This method did not appear to employ a halide.

Attia (U.S. Patent 65080281) described a sorbent for mercury and a variety of other contaminants comprising an inorganic aerogel composition. This method did not appear to employ a carbon support or halide.

Following is a List of Various Background References:

Lancia, A.; Musmarra, D.; Pepe, F.; Volpicelli, G. *Combust. Sci. & Technol.* **1993,** *93,* 277.

Streng, S.; Kassebohm, B. *Fuel Proc. Technol.* **1994,** *39,431.*

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U.S. Patent 6,214,304 Rosenthal

U.S. Patent 4,889,698 Moller

U.S. Patent 4,956,162 Smith

U.S. Patent 5,672,323 Bhat.

U.S. Patent 5,827,352 Altman

U.S. Patent 6,027,551 Hwang

U.S. Patent 5,505,766 Chang

U.S. Patent 5,607,496 Brooks

U.S. Patent 6,383,981 Blankenship

U.S. Patent 4,786,483 Audeh

U.S. Patent 3,194,629 Dreibelbis, et al

U.S. Patent 4,101,631 Ambrsini, et al

U.S. Patent 3,662,523 Revoir

U.S. Patent 4,708,853 Matviya

U.S. Patent 6,258,334 Gadkaree

U.S. Patent 6,136,072 Sjostrom

U.S. Patent Application 2002/0150516

U.S. Patent Application 2001/0003116 Neufert

CLAIMS

1. A method for separating mercury from a mercury-containing gas, the method comprising:

combusting a mixture of coal and an additive, to form the mercury-containing gas, wherein the additive is chosen from halides, halogens, salts thereof, and combinations thereof;

adding a particulate sorbent material comprising activated carbon into the mercury-containing gas;

promoting at least a portion of the sorbent material by chemically reacting the sorbent material with the additive in the mercury-containing gas, to form a promoted sorbent material;

chemically reacting mercury in the mercury-containing gas with the promoted sorbent material, to form a mercury-sorbent; and

separating the mercury-sorbent from the mercury-containing gas.

2. The method of claim 1, further comprising mixing the additive and the coal before combusting the coal.

3. The method of claim 2, wherein the additive mixed with the coal is in a liquid.

4. The method of claim 3, wherein the liquid is a hydrocarbon, a chlorinated hydrocarbon, supercritical carbon dioxide, water, salt solution, or a combination thereof.

5. The method of claim 1, wherein the promoted sorbent material comprises active sites, wherein the active sites comprise halide anions bound to the activated carbon.

6. The method of claim 1, wherein the promoted sorbent material comprises active sites, wherein the active sites comprise carbocations bound to halide anions.

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7. The method of claim 1, wherein chemically reacting the sorbent material with the additive in the mercury-containing gas comprises chemically reacting carbene species edge sites in the activated carbon with the additive.

8. The method of claim 1, wherein carbocations in the activated carbon of the mercury-sorbent accept electrons from mercury atoms of the mercury-sorbent.

9. The method of claim 1, wherein the additive is substantially in vapor form in the mercury-containing gas.

10. The method of claim 1, wherein the additive is chosen from Cl_2, HCl, sodium chloride, calcium chloride, and a combination thereof.

11. The method of claim 1, wherein the additive is chosen from Br_2, HBr, sodium bromide, calcium bromide, and a combination thereof.

12. The method of claim 1, wherein the additive is chosen from PBr_3, SCl_2, and a combination thereof.

13. The method of claim 1, wherein the additive is chosen from I_2, HI, sodium iodide, calcium iodide, potassium iodide, and combinations thereof.

14. The method of claim 1, wherein the activated carbon is chosen from powdered activated carbon, granular activated carbon, carbon black, carbon fiber, aerogel carbon, pyrolysis char, and combinations thereof.

15. The method of claim 1, further comprising adding an alkaline component to the mercury-containing gas, the alkaline component chosen from alkali elements, alkaline earth elements, alkali salts, alkaline earth salts, and combinations thereof.

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16. The method of claim 1, further comprising contacting at least one of the sorbent material and the promoted sorbent material with a mercury-stabilizing reagent chosen from S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof.

17. The method of claim 1, further comprising contacting at least one of the sorbent material and the promoted sorbent material with a secondary component chosen from halogens, hydrogen halides, Group V halides, Group VI halides, and combinations thereof.

18. The method of claim 17, wherein the secondary component is chosen from halogens, halide salts, HI, HBr, HCl, and combinations thereof.

19. The method of claim 17, wherein the secondary component is chosen from I_2, HI, and combinations thereof.

20. A method for separating mercury from a mercury-containing gas, the method comprising:

combusting coal mixed with an additive, wherein the additive is chosen from halides, halogens, salts thereof, and combinations thereof, to form the mercury-containing gas;

injecting a sorbent material comprising activated carbon into the mercury-containing gas;

promoting at least a portion of the sorbent material by chemically reacting the sorbent material with the additive in the mercury-containing gas, to form a promoted sorbent material;

chemically reacting mercury in the mercury-containing gas with the promoted sorbent material, to form a mercury-sorbent; and

separating the mercury-sorbent from the mercury-containing gas;

wherein the coal and promoter are mixed at a location upstream of a combustion furnace where combustion occurs.

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Abstract

A promoted activated carbon sorbent is described that is highly effective for the removal of mercury from flue gas streams. The sorbent comprises a new modified carbon form containing reactive forms of halogen and halides. Optional components may be added to increase reactivity and mercury capacity. These may be added directly with the sorbent, or to the flue gas to enhance sorbent performance and/or mercury capture. Mercury removal efficiencies obtained exceed conventional methods. The sorbent can be regenerated and reused. Sorbent treatment and preparation methods are also described. New methods for in-flight preparation, introduction, and control of the active sorbent into the mercury contaminated gas stream are described.

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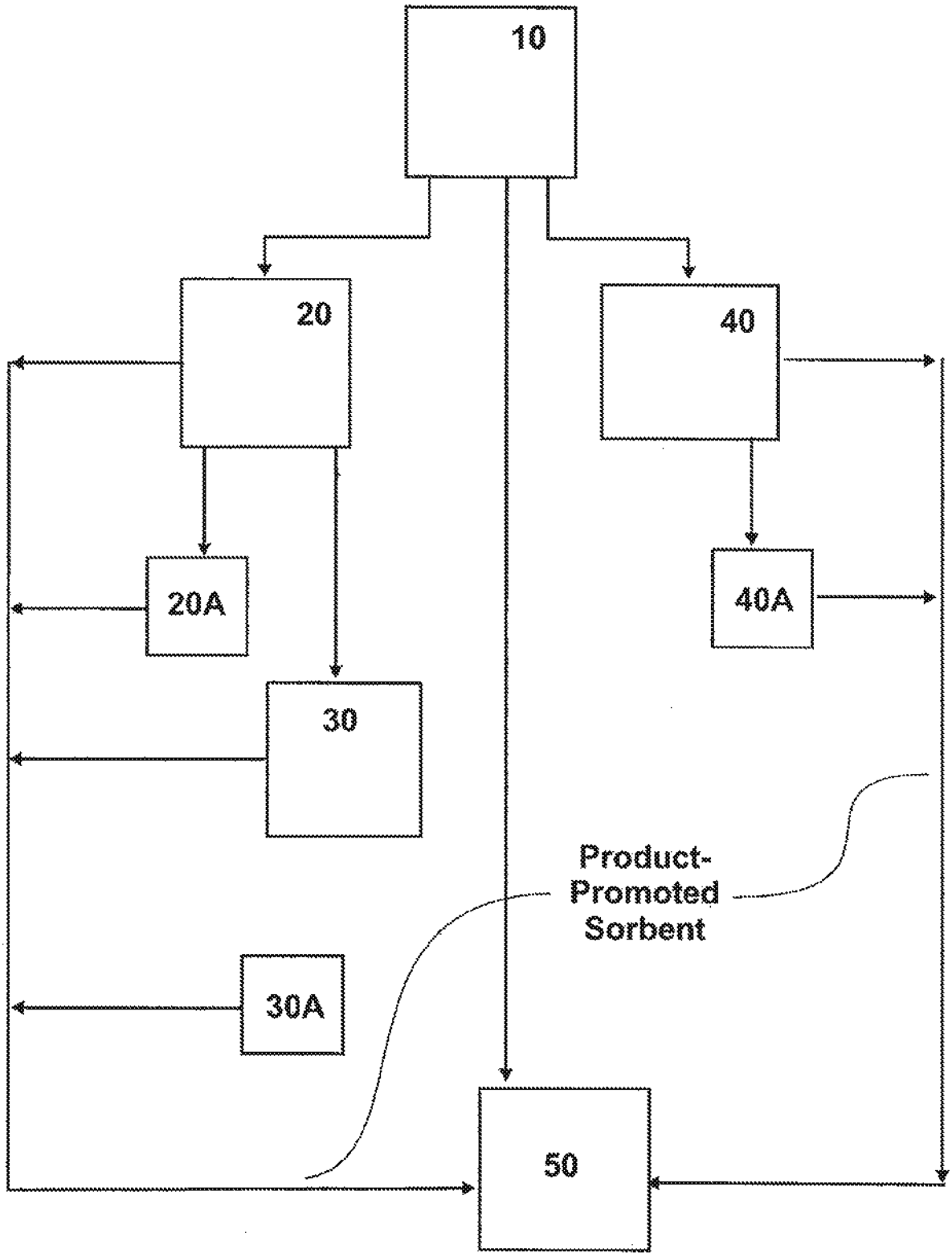


FIG. 1

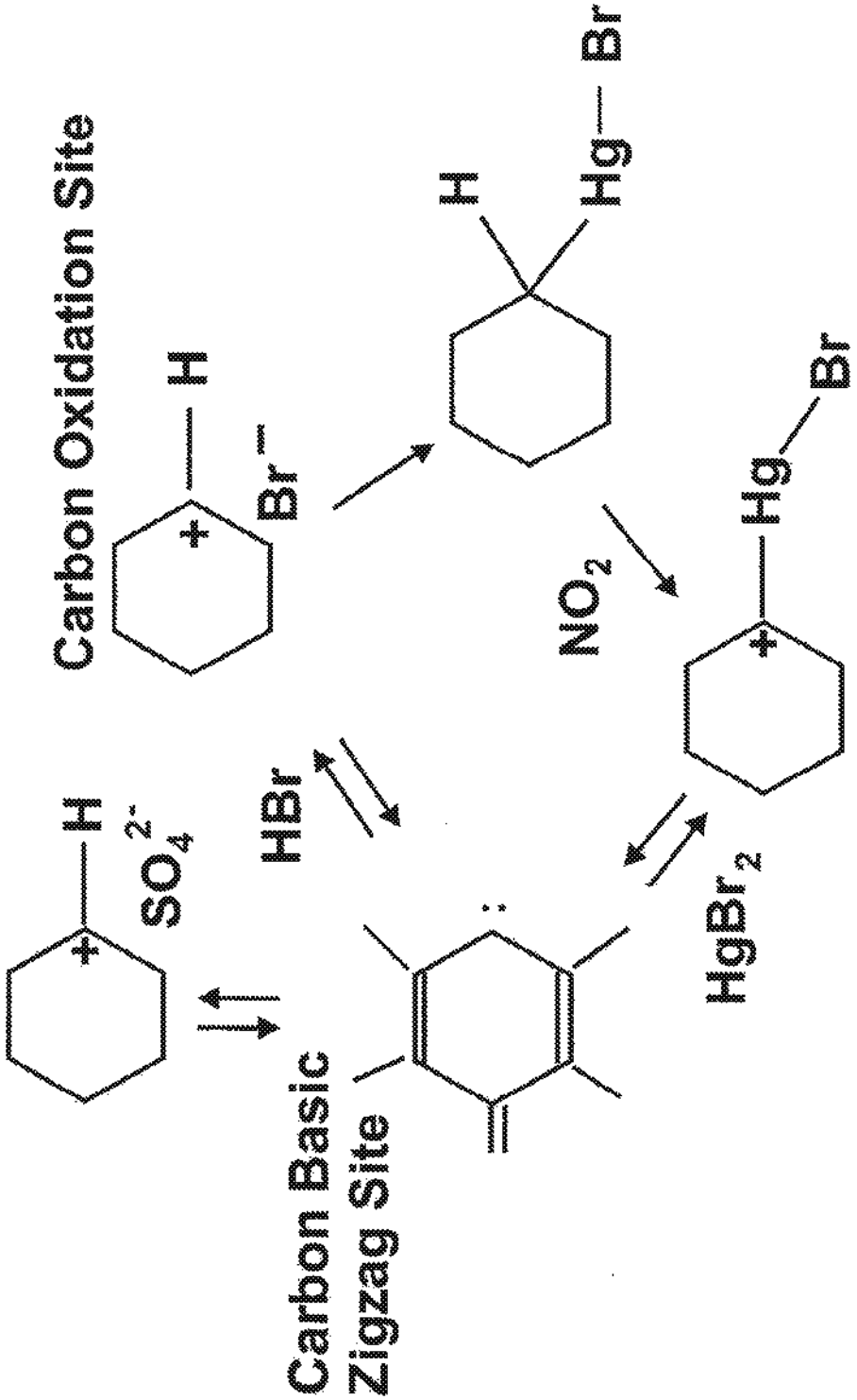

Carbon Oxidation Site
H
Br⁻
H
Hg—Br
NO2
Hg
Br
HBr
HgBr2
H
SO4 2-
Carbon Basic
Zigzag Site

FIG. 2

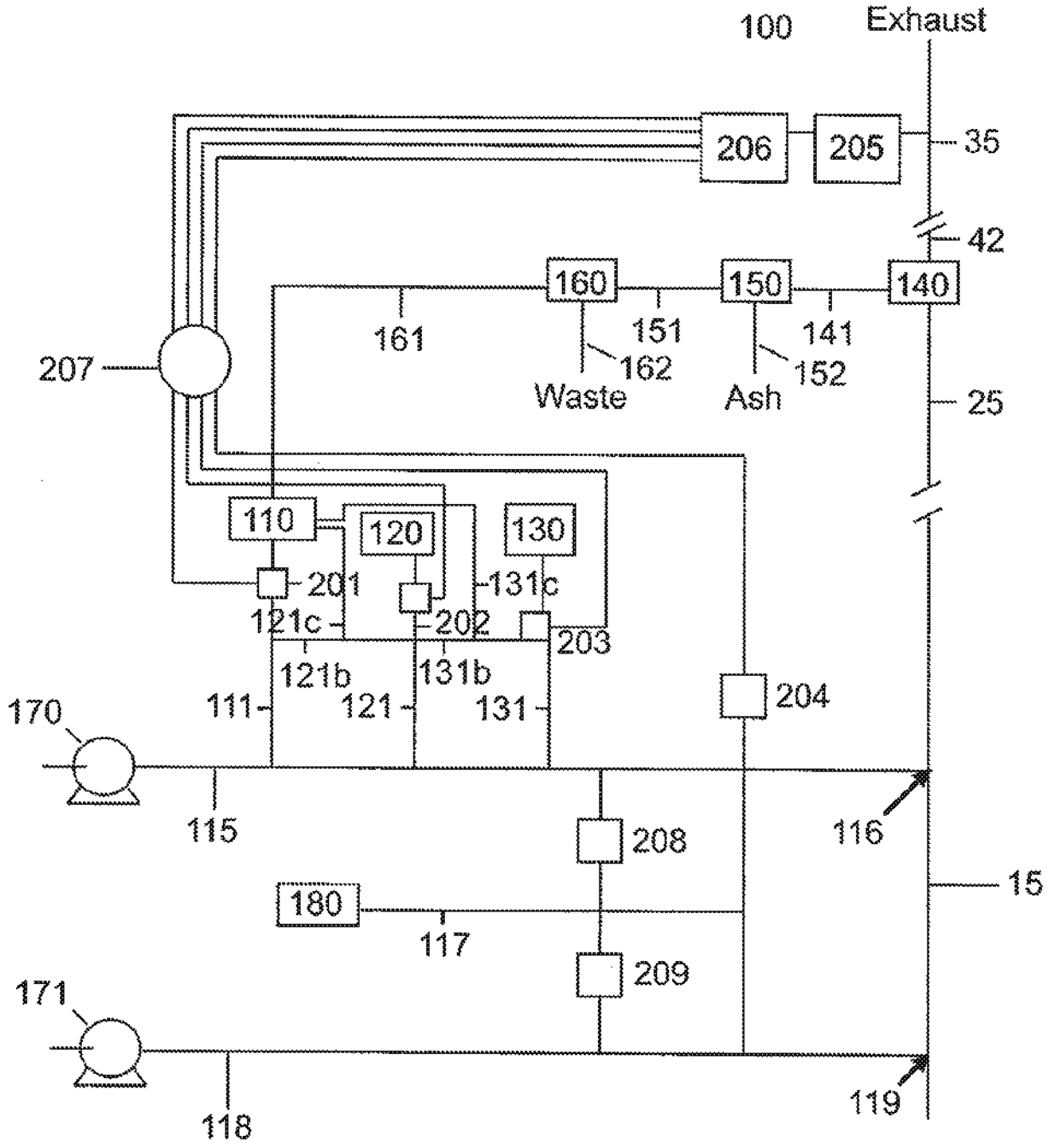

100
Exhaust
206
205
35
42
160
150
140
161
151
141
162
152
207
Waste
Ash
25
110
120
130
201
131c
121c
202
203
121b
131b
111
121
131
204
170
115
208
116
15
180
117
209
171
118
119

FIG. 3

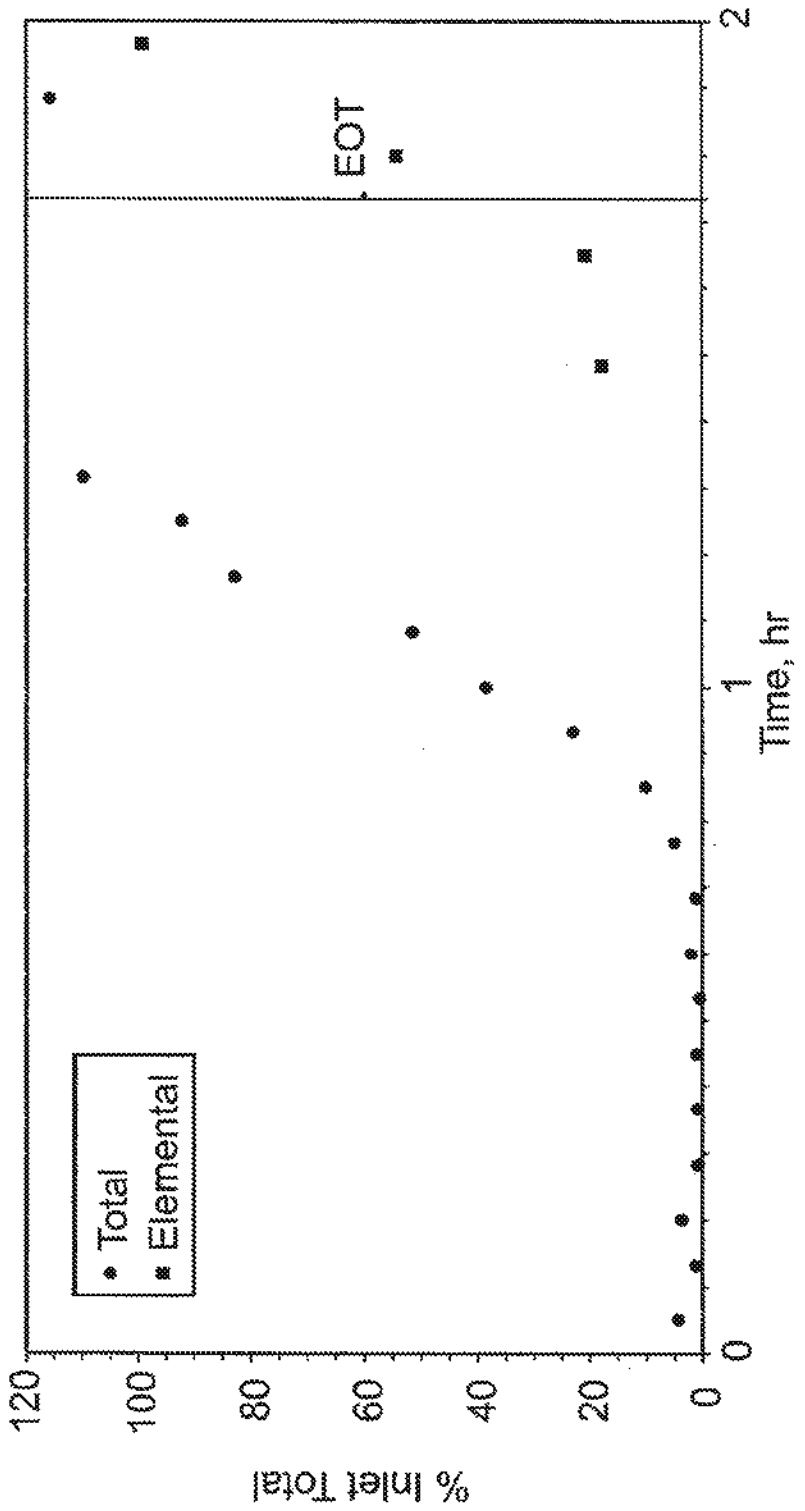

% Inlet Total
120
100
80
60
40
20
0
0
1
2
Time, hr
EOT
Total
Elemental

FIG. 4

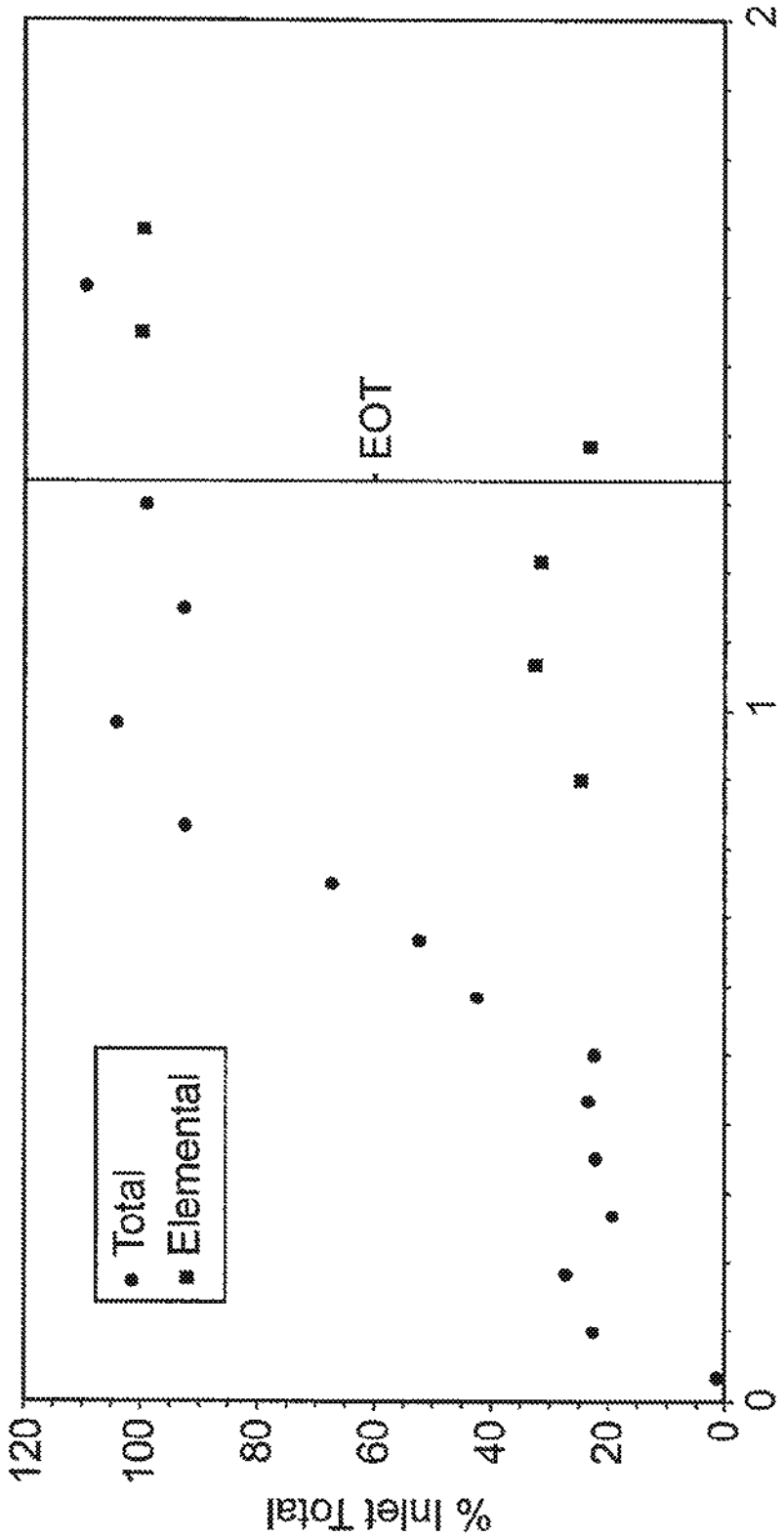

% Inlet Total
120
100
80
60
40
20
0
0
1
2
Time, hr
EOT
Total
Elemental

FIG. 5

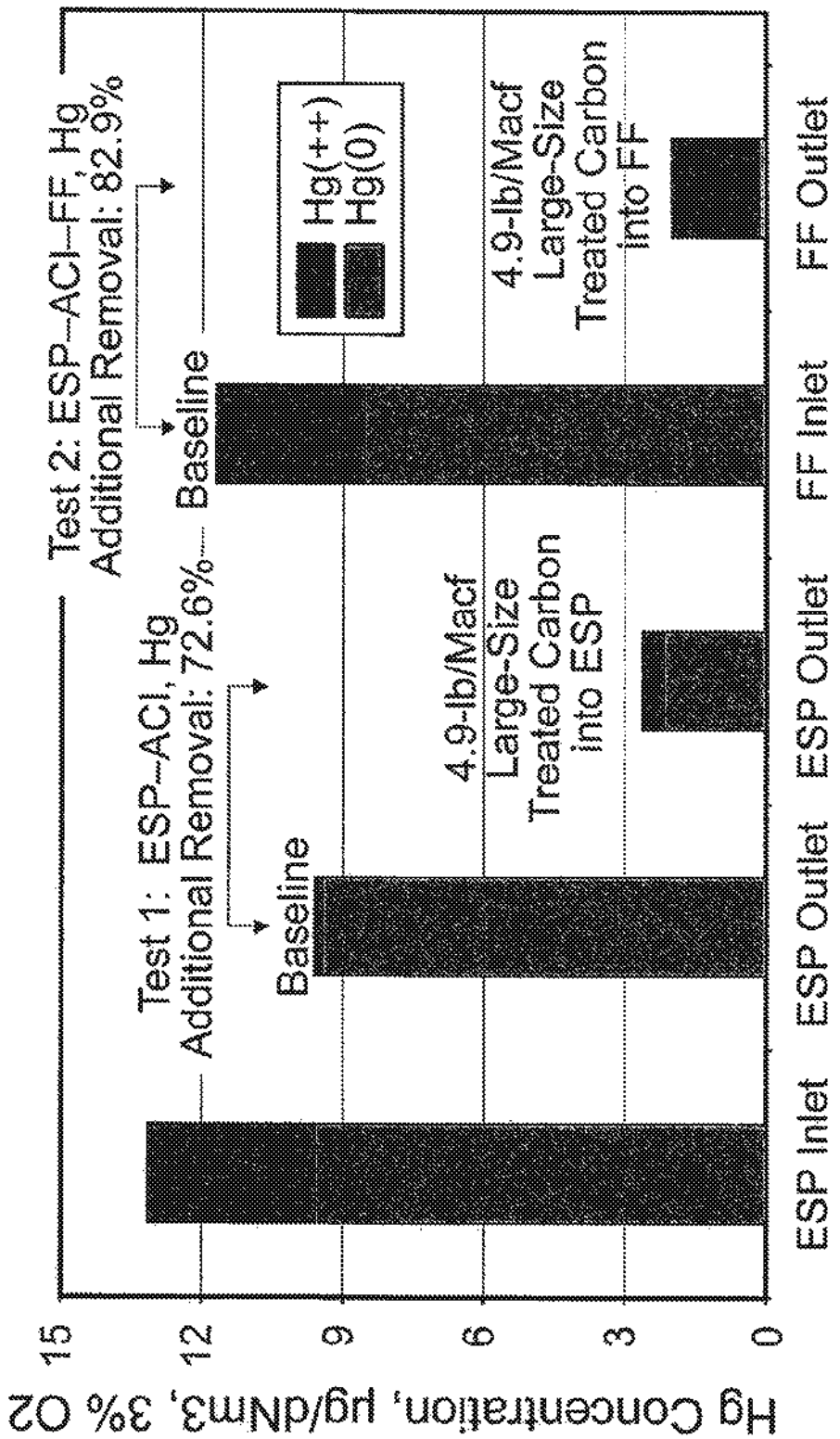

Hg Concentration, µg/dNm3, 3% O2
15
12
9
6
3
0
ESP Inlet
ESP Outlet
ESP Outlet
FF Inlet
FF Outlet
Test 1: ESP–ACI, Hg
Additional Removal: 72.6%
Baseline
4.9-lb/Macf
Large-Size
Treated Carbon
into ESP
Test 2: ESP–ACI–FF, Hg
Additional Removal: 82.9%
Baseline
4.9-lb/Macf
Large-Size
Treated Carbon
into FF
Hg(++)
Hg(0)

FIG. 6

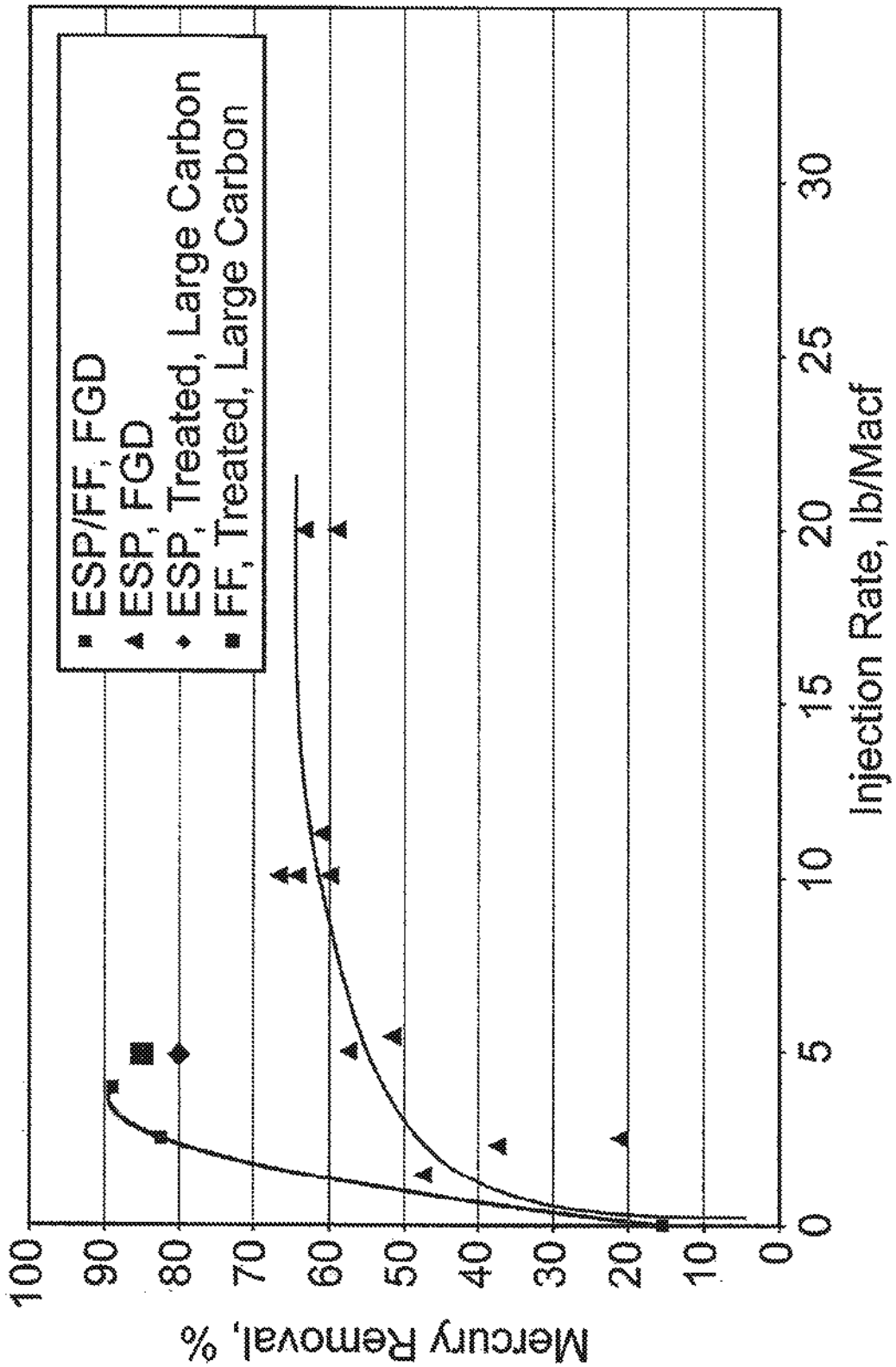

ESP/FF, FGD
ESP, FGD
ESP, Treated, Large Carbon
FF, Treated, Large Carbon
Mercury Removal, %
100
90
80
70
60
50
40
30
20
10
0
0
5
10
15
20
25
30
Injection Rate, lb/Macf

FIG. 7

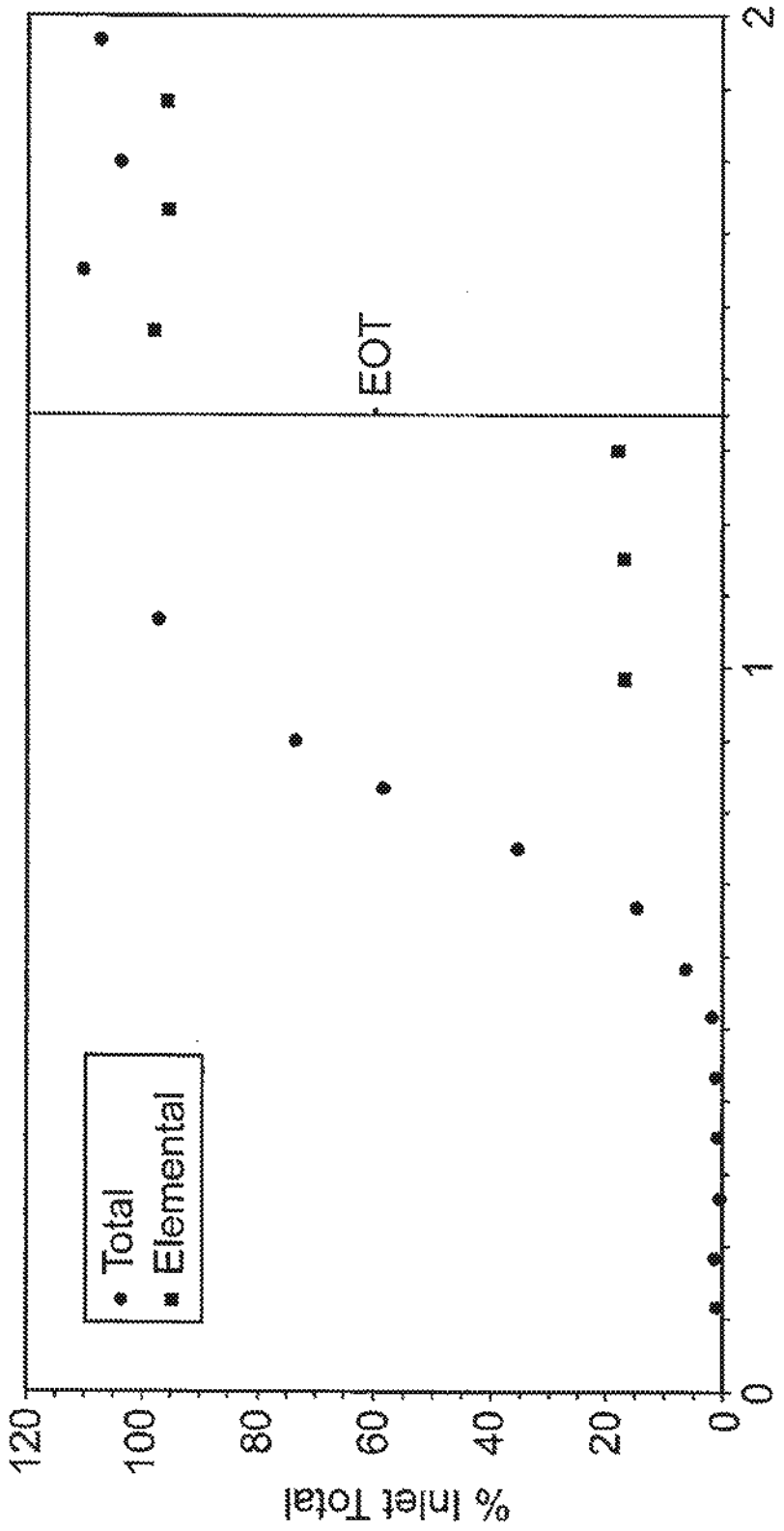

% Inlet Total
120
100
80
60
40
20
0
0
1
2
Time, hr
EOT
Total
Elemental

FIG. 8

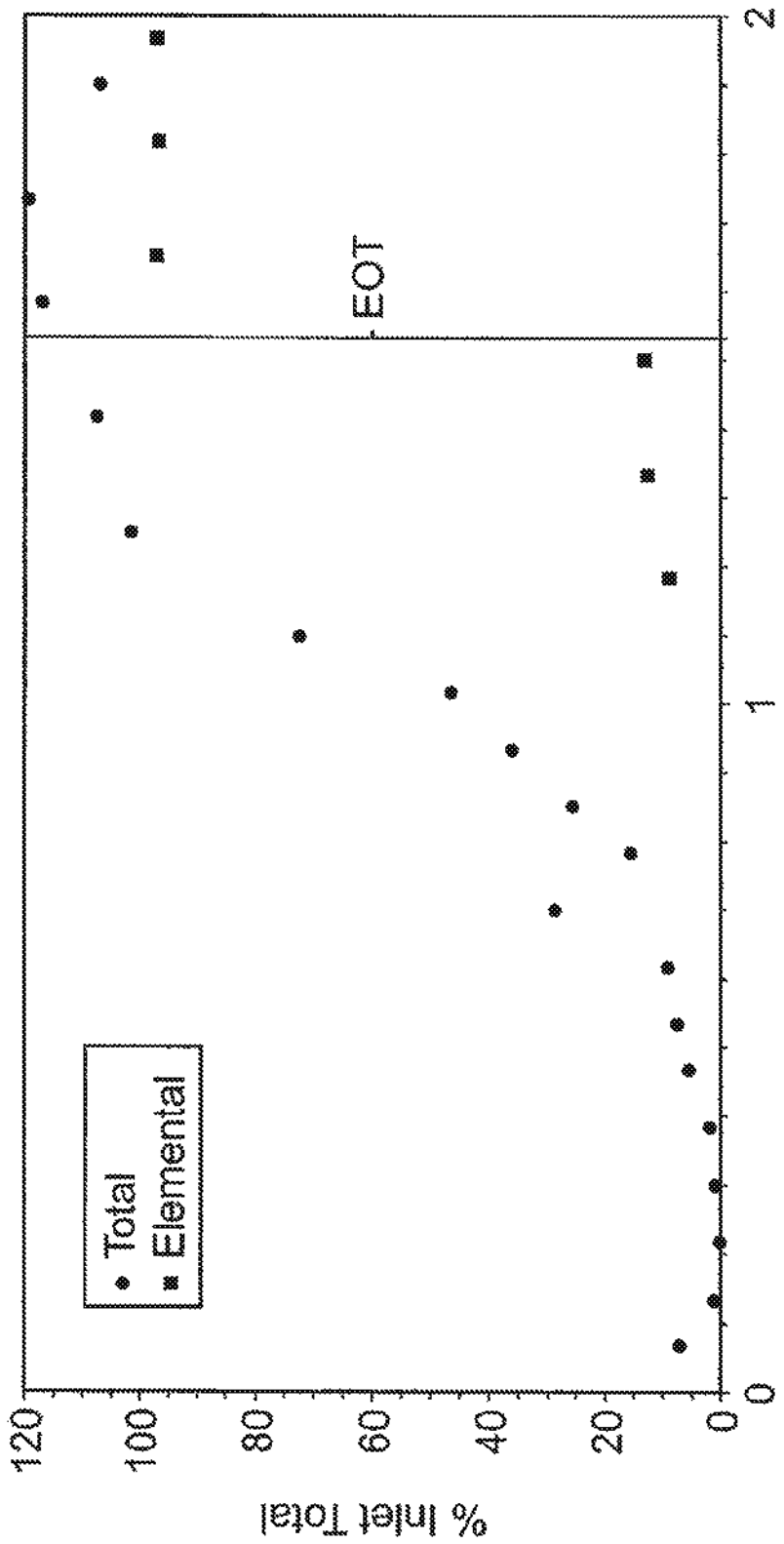

% Inlet Total
120
100
80
60
40
20
0
0
1
2
Time, hr
EOT
Total
Elemental

FIG. 9

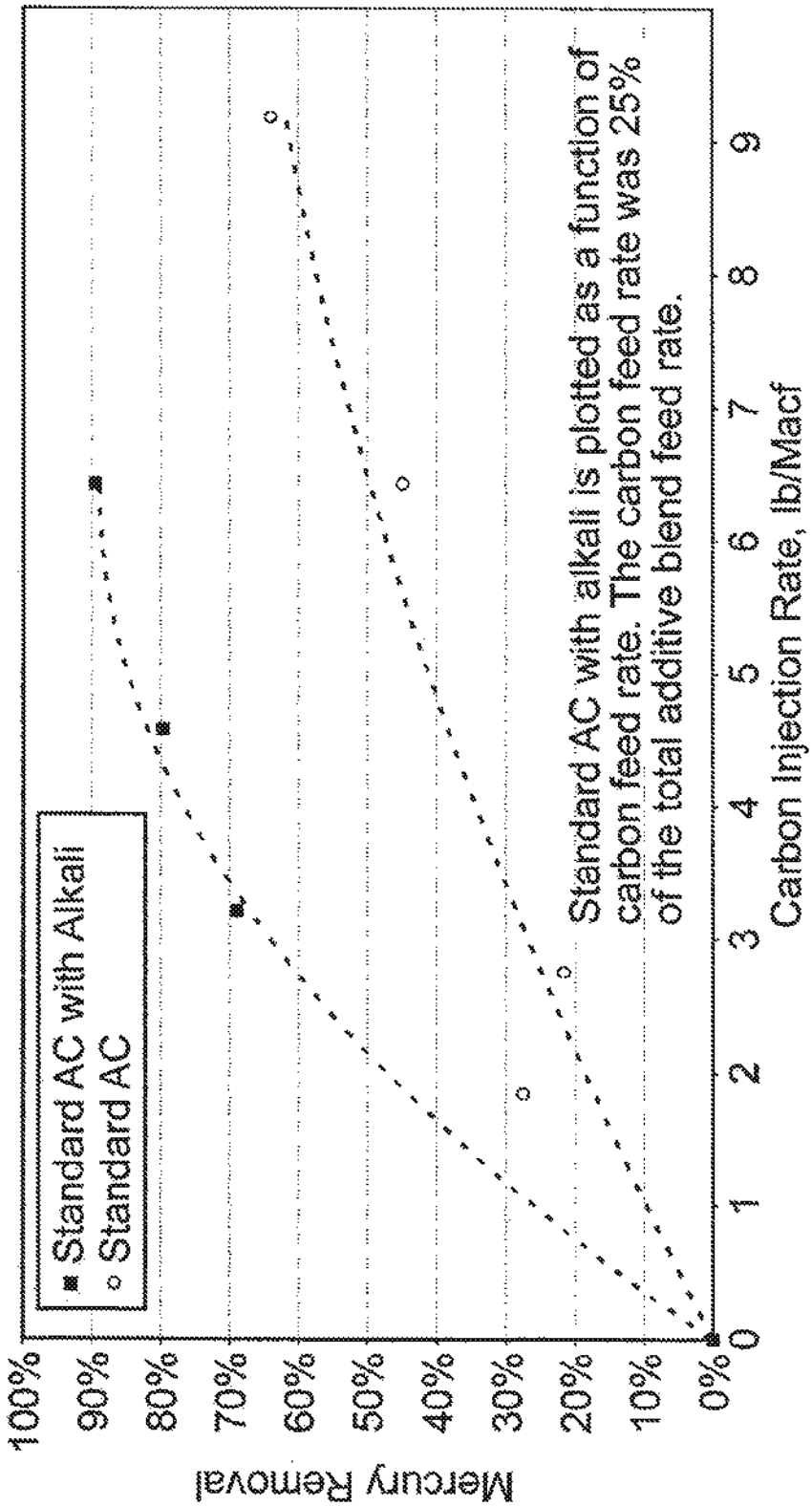

Mercury Removal
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
0
1
2
3
4
5
6
7
8
9
Carbon Injection Rate, lb/Macf
Standard AC with Alkali
Standard AC
Standard AC with alkali is plotted as a function of carbon feed rate. The carbon feed rate was 25% of the total additive blend feed rate.

FIG. 10

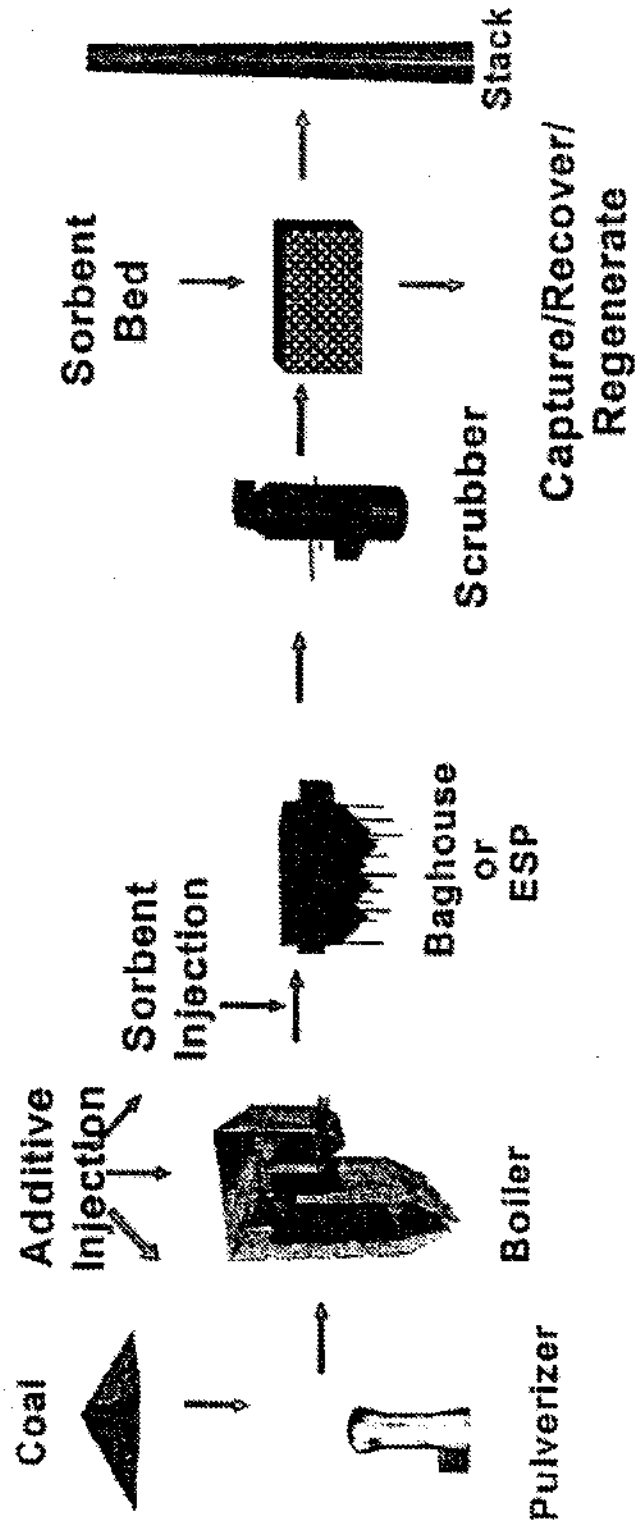

Coal
Pulverizer
Additive Injection
Boiler
Sorbent Injection
Baghouse or ESP
Scrubber
Capture/Recover/ Regenerate
Sorbent Bed
Stack

FIG. 11

Electronic Patent Application Fee Transmittal

Application Number:	
Filing Date:	
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Filer:	Jose M. Nunez/Megan Reed
Attorney Docket Number:	4820.001US8

Filed as Large Entity

Filing Fees for Utility under 35 USC 111(a)

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

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
Extension-of-Time:				
Miscellaneous:				
		Total in USD ($)		**1880**

Electronic Acknowledgement Receipt

EFS ID:	32327251
Application Number:	15951970
International Application Number:	
Confirmation Number:	6410
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Customer Number:	21186
Filer:	Jose M. Nunez/Megan Reed
Filer Authorized By:	Jose M. Nunez
Attorney Docket Number:	4820.001US8
Receipt Date:	12-APR-2018
Filing Date:	
Time Stamp:	17:46:10
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	DA
Payment was successfully received in RAM	$1880
RAM confirmation Number	041318INTEFSW00004490190743
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File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1		SIGNED_4820_001US8_Application.pdf	3657154 1676f2fe70da8780579d52e610e67d0858e3fa8b	yes	81

Multipart Description/PDF files in .zip description

Document Description	Start	End
Transmittal of New Application	1	1
Application Data Sheet	2	9
Specification	10	66
Claims	67	69
Abstract	70	70
Drawings-only black and white line drawings	71	81

Warnings:

Information:

2	Fee Worksheet (SB06)	fee-info.pdf	36718 13a94b5000cac4e637fc705efee2a9cdd31067e2	no	2

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Information:

Total Files Size (in bytes):	3693872

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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.

Electronic Acknowledgement Receipt

EFS ID:	33295984
Application Number:	15951970
International Application Number:	
Confirmation Number:	6410
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Customer Number:	21186
Filer:	Jordan Rivard Golomb/Jamie Johnson
Filer Authorized By:	Jordan Rivard Golomb
Attorney Docket Number:	4820.001US8
Receipt Date:	26-JUL-2018
Filing Date:	12-APR-2018
Time Stamp:	16:47:32
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

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1	Non Patent Literature	0297_dunham.pdf	2906300 999f74876108ac44945ebe92a2290bf0fced624e	no	6

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3	Non Patent Literature	0302_ghorishi_1998.pdf	5713537 a44d307431b796a5351b222702ba6f9b34d 48c0a	no	12
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9	Non Patent Literature	0308_kyotani_1988.pdf	614835 6488fd6f91b7e03b645380faa5aebf4b540d3e62a	no	3
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10	Non Patent Literature	0309_lancia_etal_1993.pdf	4418191 e329cf899781ccaa0b706bddd65da7a3a1082c7e	no	13
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17	Non Patent Literature	0316_nguyen_thanh_bandosz_2006.pdf	1275237 1952e36c32ee32f7ea67bf7674736beb12899159	no	9
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19	Non Patent Literature	0318_3338013us1_olson_reduction_111808.pdf	2211800 eef96dd525a025775c67aaee77787aef93230fcc	no	13
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27	Non Patent Literature	0326_3338013us1_streng_toxics_111808.pdf	3793209 e943d53c31c460472f7b9f75f28fec4067e9c9b7	no	14
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34	Non Patent Literature	0333_wang_smith.pdf	949889 4ec97da19e86c7abb7ab861a208305271ba693cd	no	8
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35	Non Patent Literature	0334_zhang_2009.pdf	6581725 37c602e4e11fd1f0c05537faeaff860ac58ed192	no	6
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36	Non Patent Literature	0335_zhao_2006.pdf	3949197 a831dd6e9cb6e648f4d58176ff194d10a1ca79a4	no	8
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37	Non Patent Literature	0336_zheng_2012.pdf	22977549 5f5979f5d42129220d87a4704b9770befa040213	no	31
Warnings:					
Information:					
38	Non Patent Literature	0337_zhu_enhanced.pdf	927796 12ca6d9cc44513140a1c580073adf9b2f60bd93e	no	7
Warnings:					
Information:					
39	Non Patent Literature	0338_3338016US2_PAMD_12-10-14_1.pdf	421845 69110a188624167ea422ffdefbb31df7c00c5244	no	6
Warnings:					
Information:					
40	Non Patent Literature	0339_jp_49_53594.pdf	242109 1c77d4fa0cb50819a54aa20b5b8c9f1b1ef06d6b	no	1
Warnings:					
Information:					
41	Non Patent Literature	0440_olson_2001.pdf	2329142 9e62423a94f3bf97ff6d8e220b90a5d8fc2e699e	no	19
Warnings:					
Information:					
42	Foreign Reference	0019_de_202012003747u1.pdf	9407344 a480a109b566cc54647793c3611c0c04eef9cdd4	no	51
Warnings:					
Information:					
43	Non Patent Literature	0298_eisazadeh_2007.pdf	277323 b0e52eba463daca0c00c8e16f497487b89669e0e	no	6
Warnings:					
Information:					

44	Non Patent Literature	0299_felsvang_etal_2001.pdf	6241839 9da7091271a0ebe0f35c2797684184e205758785	no	19
Warnings:					
Information:					
45	Non Patent Literature	0300_fernandez_saavedra_2008.pdf	5391004 5902c66a11bfe07fd569c873c887cda23401a58f	no	10
Warnings:					
Information:					
		Total Files Size (in bytes):	167050927		

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.

PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference	FOR FURTHER ACTION	see Form PCT/ISA/220 as well as, where applicable, item 5 below.
2380-01102		
International application No.	International filing date *(day/month/year)*	(Earliest) Priority Date *(day/month/year)*
PCT/US2008/071986	**01 AUGUST 2008 (01.08.2008)**	01 AUGUST 2007 (01.08.2007)

Applicant

ENERGY & ENVIRONMENTAL RESEARCH CENTER et al

This International search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 3 sheets.

☐ It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**
   a. With regard to the **language**, the international search was carried out on the basis of :
      ☒ the international application in the language in which it was filed
      ☐ a translation of the international application into _____, which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b))
   b. ☐ This international search report has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43.6*bis*(a)).
   c. ☐ With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, see Box No. I.
2. ☐ **Certain claims were found unsearchable** (See Box No. II)
3. ☐ **Unity of invention is lacking** (See Box No. III)
4. With regard to the **title**,
   ☒ the text is approved as submitted by the applicant.
   ☐ the text has been established by this Authority to read as follows:
5. With regard to the **abstract**,
   ☒ the text is approved as submitted by the applicant.
   ☐ the text has been established, according to Rule 38.2, by this Authority as it appears in Box No. IV. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.
6. With regard to the drawings,
   a. the figure of the drawings to be published with the abstract is Figure No. 1
      ☒ as suggested by the applicant.
      ☐ because the applicant failed to suggest a figure.
      ☐ because this figure better characterizes the invention.
   b. ☐ none of the figure is to be published with the abstract.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US2008/071986

A. CLASSIFICATION OF SUBJECT MATTER

F17C 5/00(2006.01)i, B01D 53/00(2006.01)i, F02C 6/14(2006.01)i, E21B 21/00(2006.01)i

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: F17C, F02C, C10G

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Korean Utility models and applications for Utility models since 1975
Japanese Utility models and applications for Utility models since 1975

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

eKIPASS (KIPO internal) & keywords: "gas compressor", "gas turbine", "sulfur", "water" and "separate"

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP 09-256812 A (TOSHIBA CORP.) 30 September 1997 See the abstract and Claim 1.	1, 10
A	US 4,342,192 (KLAUS HEYN et al.) 3 August 1982 See the abstract and Fig. 1.	1, 10
A	US 7,017,330 B2 (JAMES C. BELLOWS) 28 March 2006 See the abstract and Fig. 1.	1, 10
A	KR 10-2002-0052209 A (JGC CORPORATION) 2 July 2002 See page 2, line 31 - page 6, line 1 and Fig. 3.	1, 10

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents,such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
30 JANUARY 2009 (30.01.2009)	**30 JANUARY 2009 (30.01.2009)**

Name and mailing address of the ISA/KR	Authorized officer
Korean Intellectual Property Office Government Complex-Daejeon, 139 Seonsa-ro, Seo-gu, Daejeon 302-701, Republic of Korea	HAN, JAE SEOB
Facsimile No. 82-42-472-7140	Telephone No. 82-42-481-5439

Form PCT/ISA/210 (second sheet) (July 2008)

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.
PCT/US2008/071986

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
JP 09-256812 A	30.09.1997	NONE	
US 4342192	03.08.1982	CA 1134219 A1	26.10.1982
		DE 2850005 A1	29.05.1980
		DE 2931427 A1	19.02.1981
US 7017330 B2	28.03.2006	US 6804964	19.10.2004
		US 7431757	07.10.2008
		US 2004-0055309 A1	25.03.2004
		US 2005-039462 A1	24.02.2005
		US 6804964 B2	19.10.2004
KR 10-2002-0052209 A	02.07.2002	DE 60002602 D1	12.06.2003
		DE 60002602 T2	19.02.2004
		EP 1230323 B1	07.05.2003
		EP 1230323 A1	14.08.2002
		ES 2195942 T3	16.12.2003
		JP 2001-140656 A	22.05.2001
		TW 477812 B	01.03.2002
		TW 477812 A	01.03.2002
		US 6786050 B1	07.09.2004
		WO 01-36566 A1	25.05.2001

PATENT COOPERATION TREATY

PCT

RECD 29 JAN 2007
WIPO PCT

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference 2380-00201	FOR FURTHER ACTION	See Form PCT/IPEA/416
International application No. PCT/US05/30018	International filing date *(day/month/year)* 23 August 2005 (23.08.2005)	Priority date *(day/month/year)* 30 August 2004 (30.08.2004)

International Patent Classification (IPC) or national classification and IPC

IPC: **B01D 53/02**(2006.01)
USPC: 95/13,107,134

Applicant

ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION

1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 3 sheets, including this cover sheet.
3. This report is also accompanied by ANNEXES, comprising:
   a. ☒ *(sent to the applicant and to the International Bureau)* a total of 5 sheets, as follows:
      ☐ sheets of the description, claims and/or drawings which have been amended and are the basis of this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions).
      ☐ sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box.
   b. ☐ *(sent to the International Bureau only)* a total of (indicate type and number of electronic carrier(s)) _____ , containing a sequence listing and/or tables related thereto, in electronic form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).
4. This report contains indications relating to the following items:
   - ☒ Box No. I Basis of the report
   - ☐ Box No. II Priority
   - ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - ☐ Box No. IV Lack of unity of invention
   - ☒ Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
   - ☐ Box No. VI Certain documents cited
   - ☐ Box No. VII Certain defects in the international application
   - ☐ Box No. VIII Certain observations on the international application

Date of submission of the demand	Date of completion of this report
25 October 2006 (25.10.2006)	27 December 2006 (27.12.2006)
Name and mailing address of the IPEA/ US Mail Stop PCT, Attn: IPEA/US Commissioner for Patents P.O. Box 1450 Alexandria, Virginia 22313-1450 Facsimile No. (571) 273-3201	Authorized officer Robert A. Hopkins Telephone No. 571-272-1700

DEBORAH A. THOMAS
PARALEGAL SPECIALIST

Form PCT/IPEA/409 (cover sheet)(April 2005)

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.
PCT/US05/30018

Box No. I Basis of the report

1. With regard to the **language**, this report is based on:

☒ the international application in the language in which it was filed.

☐ a translation of the international application into _____, which is the language of a translation furnished for the purposes of:

- ☐ international search (under Rules 12.3 and 23.1(b))
- ☐ publication of the international application (under Rule 12.4(a))
- ☐ international preliminary examination (under Rules 55.2(a) and/or 55.3(a))

2. With regard to the **elements** of the international application, this report is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report)*:

☐ the international application as originally filed/furnished

☒ the description:
pages 1-28 as originally filed/furnished
pages* NONE received by this Authority on _____
pages* NONE received by this Authority on _____

☒ the claims:
pages NONE as originally filed/furnished
pages* NONE as amended (together with any statement) under Article 19
pages* 29-33 received by this Authority on 25 October 2006 (25.10.2006)
pages* NONE received by this Authority on _____

☒ the drawings:
pages 1-7 as originally filed/furnished
pages* NONE received by this Authority on _____
pages* NONE received by this Authority on _____

☐ a sequence listing and/or any related table(s) - see Supplemental Box Relating to Sequence Listing.

3. ☒ The amendments have resulted in the cancellation of:

- ☒ the description, pages NONE
- ☒ the claims, Nos. 48-54
- ☒ the drawings, sheets/figs NONE
- ☐ the sequence listing (*specify*): _____
- ☐ any table(s) related to the sequence listing (*specify*): _____

4. ☐ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).

- ☐ the description, pages _____
- ☐ the claims, Nos. _____
- ☐ the drawings, sheets/figs _____
- ☐ the sequence listing (*specify*): _____
- ☐ any table(s) related to the sequence listing (*specify*): _____

** If item 4 applies, some or all of those sheets may be marked "superseded."*

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.
PCT/US05/30018

Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Claims 34-47	YES
	Claims 1-33	NO
Inventive Step (IS)	Claims 34-47	YES
	Claims 1-33	NO
Industrial Applicability (IA)	Claims 1-47	YES
	Claims NONE	NO

2. Citations and Explanations (Rule 70.7)

Claims 1-33 lack novelty under PCT Article 33(2) as being anticipated by NISHINO ET AL. Note the base activated carbon and impregnated halide or halogen. Note also the optional secondary component including an alkaline earty salt. Note also the use of an organic solvent(ethanol). Note also the activated carbon with promoter is used for removal of mercury vapor. Note also the sorbent is either a fixed bed, moving bed, or fluidized bed sorbent. Note also the mercury content of the flue gas is monitored, and the sorbetn is recovered from the clean gas. Examiner also notes that the amendment "wherein the composition of the promoted carbon sorbent is controlled by the mercury content of the cleaned gas stream" does not have patentable weight upon a product claim(claim 1) and a method of making a product claim(claim 17)

Claims 1 and 17 lack novelty under PCT Article 33(2) as being anticipated by REVOIR ET AL. REVOIR ET AL teaches a base activated carbon and impregnated halide or halogen. Examiner also notes that the amendment "wherein the composition of the promoted carbon sorbent is controlled by the mercury content of the cleaned gas stream" does not have patentable weight upon a product claim(claim 1) and a method of making a product claim(claim 17).

Claims 1-47 meet the criteria set out in PCT Article 33(4), and thus have industrial applicability because the subject matter claimed can be made or used in industry.

Claims 34-47 meet the criteria set out in PCT Article 33(2)-(3), because the prior art does not teach or fairly suggest a step of preparing the sorbent by in-flight reaction of the base activated carbon with the at least one component within a transport line from which the reaction product is injected into the mercury containing gas stream.

---------- NEW CITATIONS ----------

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US05/30018

A. CLASSIFICATION OF SUBJECT MATTER
IPC: **B01D 53/02**(2006.01)

USPC: 95/13,107,134
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)
U.S. : 95/13,107,134

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
NONE

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
Please See Continuation Sheet

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 4,500,327 A (NISHINO et al) 19 February 1985, column 1 lines 66-68, column 2 lines 9-17, column 2 lines 34-38.	1-54
X	US 3,662,523A (REVOIR et al) 16 May 1972, column 1 lines 5-10, lines 62-69, column 2 lines 1-11.	1,16

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent 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
12 June 2006 (12.06.2006)	25 JUL 2006
Name and mailing address of the ISA/US Mail Stop PCT, Attn: ISA/US Commissioner for Patents P.O. Box 1450 Alexandria, Virginia 22313-1450 Facsimile No. (571) 273-3201	Authorized officer Robert A. Hopkins Telephone No. 703-872-9306

Form PCT/ISA/210 (second sheet) (April 2005)

Continuation of B. FIELDS SEARCHED Item 3:
EAST

search terms: activated adj carbon, halide, halogen, mercury, sorbent, adsorption , adsorb

PATENT COOPERATION TREATY

REC'D 28 JUL 2006
PCT

From the
INTERNATIONAL SEARCHING AUTHORITY

To:
TIMOTHY S. WESTBY
CONLEY ROSE, P.C.
P.O. BOX 3267
HOUSTON, TX 77253-3267

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

(PCT Rule 43*bis*.1)

Date of mailing (*day/month/year*)	25 JUL 2006
Applicant's or agent's file reference 2380-00201	**FOR FURTHER ACTION** See paragraph 2 below

International application No.	International filing date (*day/month/year*)	Priority date (*day/month/year*)
PCT/US05/30018	23 August 2005 (23.08.2005)	30 August 2004 (30.08.2004)

International Patent Classification (IPC) or both national classification and IPC

IPC: **B01D 53/02**(2006.01)
USPC: 95/13,107,134

Applicant

ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION

1. This opinion contains indications relating to the following items:

- [X] Box No. I — Basis of the opinion
- [ ] Box No. II — Priority
- [ ] Box No. III — Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- [ ] Box No. IV — Lack of unity of invention
- [X] Box No. V — Reasoned statement under Rule 43*bis*.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- [ ] Box No. VI — Certain documents cited
- [ ] Box No. VII — Certain defects in the international application
- [ ] Box No. VIII — Certain observations on the international application

2. **FURTHER ACTION**

If a demand for international preliminary examination is made, this opinion will be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA") except that this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1*bis*(*b*) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of 3 months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

3. For further details, see notes to Form PCT/ISA/220.

Name and mailing address of the ISA/ US	Date of completion of this opinion	Authorized officer
Mail Stop PCT, Attn: ISA/US Commissioner for Patents P.O. Box 1450 Alexandria, Virginia 22313-1450 Facsimile No. (571) 273-3201	12 June 2006 (12.06.2006)	Robert A. Hopkins Telephone No. 703-872-9306

Form PCT/ISA/237 (cover sheet) (April 2005)

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

International application No.
PCT/US05/30018

Box No. I Basis of this opinion

1. With regard to the **language**, this opinion has been established on the basis of:

☒ the international application in the language in which it was filed

☐ a translation of the international application into _____, which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b)).

2. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application and necessary to the claimed invention, this opinion has been established on the basis of:

a. type of material

☐ a sequence listing

☐ table(s) related to the sequence listing

b. format of material

☐ on paper

☐ in electronic form

c. time of filing/furnishing

☐ contained in the international application as filed.

☐ filed together with the international application in electronic form.

☐ furnished subsequently to this Authority for the purposes of search.

3. ☐ In addition, in the case that more than one version or copy of a sequence listing and/or table(s) relating thereto has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.

4. Additional comments:

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

International application No.
PCT/US05/30018

Box No. V Reasoned statement under Rule 43 *bis*.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Claims NONE	YES
	Claims 1-54	NO
Inventive step (IS)	Claims NONE	YES
	Claims 1-54	NO
Industrial applicability (IA)	Claims 1-54	YES
	Claims NONE	NO

2. Citations and explanations:

Claims 1-54 are novelty under PCT Article 33(2) as being anticipated by NISHINO ET AL. Note the base activated carbon and impregnated halide or halogen. Note also the optional secondary component including an alkaline earty salt. Note also the use of an organic solvent(ethanol). Note also the activated carbon with promoter is used for removal of mercury vapor. Note also the sorbent is either a fixed bed, moving bed, or fluidized bed sorbent. Note also the mercury content of the flue gas is monitored, and the sorbetn is recovered from the clean gas.

Claims 1 and 16 lack novelty under PCT Article 33(2) as being anticipated by REVOIR ET AL. REVOIR ET AL teaches a base activated carbon and impregnated halide or halogen.

Claims 1-54 meet the criteria set out in PCT Article 33(4), and thus have industrial applicability because the subject matter claimed can be made or used in industry.

PATENT COOPERATION TREATY

From the INTERNATIONAL BUREAU

PCT

NOTIFICATION CONCERNING TRANSMITTAL OF COPY OF INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY (CHAPTER I OF THE PATENT COOPERATION TREATY)
(PCT Rule 44bis.1(c))

To:

KOWALCHYK, Katherine, M.
Merchant & Gould P.C.
P.O. Box 2903
Minneapolis, MN 55402-0903
ETATS-UNIS D'AMERIQUE

Date of mailing *(day/month/year)*	
03 November 2011 (03.11.2011)	
Applicant's or agent's file reference	IMPORTANT NOTICE
16291.3WO01	

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/US2010/022807	02 February 2010 (02.02.2010)	23 April 2009 (23.04.2009)

Applicant
ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION et al

The International Bureau transmits herewith a copy of the international preliminary report on patentability (Chapter I of the Patent Cooperation Treaty)

The International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland

Facsimile No. +41 22 338 82 70

Authorized officer:
Lingfei Bai

e-mail: pt02.pct@wipo.int

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter I of the Patent Cooperation Treaty)

(PCT Rule 44*bis*)

Applicant's or agent's file reference 16291.3WO01	**FOR FURTHER ACTION**	See item 4 below
International application No. PCT/US2010/022607	International filing date *(day/month/year)* 02 February 2010 (02.02.2010)	Priority date *(day/month/year)* 23 April 2009 (23.04.2009)
International Patent Classification (8th edition unless older edition indicated) See relevant information in Form PCT/ISA/237		
Applicant ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION		

1. This international preliminary report on patentability (Chapter I) is issued by the International Bureau on behalf of the International Searching Authority under Rule 44 *bis*.1(a).

2. This REPORT consists of a total of 5 sheets, including this cover sheet.

   In the attached sheets, any reference to the written opinion of the International Searching Authority should be read as a reference to the international preliminary report on patentability (Chapter I) instead.

3. This report contains indications relating to the following items:

☒	Box No. I	Basis of the report
☐	Box No. II	Priority
☐	Box No. III	Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
☐	Box No. IV	Lack of unity of invention
☒	Box No. V	Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
☐	Box No. VI	Certain documents cited
☐	Box No. VII	Certain defects in the international application
☐	Box No. VIII	Certain observations on the international application

4. The International Bureau will communicate this report to designated Offices in accordance with Rules 44*bis*.3(c) and 93*bis*.1 but not, except where the applicant makes an express request under Article 23(2), before the expiration of 30 months from the priority date (Rule 44*bis* .2).

	Date of issuance of this report 25 October 2011 (25.10.2011)
The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Authorized officer Lingfei Bai
Facsimile No. +41 22 338 82 70	e-mail: pt02 pct@wipo.int

Form PCT/IB/373 (January 2004)

PATENT COOPERATION TREATY

From the
INTERNATIONAL SEARCHING AUTHORITY

To:
KOWALCHYK KATHERINE M

MERCHANT & GOULD P.C. P.O. BOX 2903
MINNEAPOLIS MN 55402-0903 USA

PCT

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

(PCT Rule 43bis.1)

Date of mailing *(day/month/year)*	**13 SEPTEMBER 2010 (13.09.2010)**
Applicant's or agent's file reference 16291.3WO11	**FOR FURTHER ACTION** See paragraph 2 below

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/US2010/022807	**02 FEBRUARY 2010 (02.02.2010)**	23 APRIL 2009 (23.04.2009)

International Patent Classification (IPC) or both national classification and IPC

B01D 53/02(2006.01)i, B01D 53/64(2006.01)i, F23J 15/00(2006.01)i

Applicant

ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION et al

1. This opinion contains indications relating to the following items:

- [x] Box No. I Basis of the opinion
- [ ] Box No. II Priority
- [ ] Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- [ ] Box No. IV Lack of unity of invention
- [x] Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- [ ] Box No. VI Certain documents cited
- [ ] Box No. VII Certain defects in the international application
- [ ] Box No. VIII Certain observations on the international application

2. **FURTHER ACTION**

If a demand for international preliminary examination is made, this opinion will be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA") except that this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1bis(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of 3 months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.
For further options, see Form PCT/ISA/220.

3. For further details, see notes to Form PCT/ISA/220.

Name and mailing address of the ISA/KR	Date of completion of this opinion	Authorized officer
Korean Intellectual Property Office Government Complex-Daejeon, 139 Seonsa-ro, Seo-gu, Daejeon 302-701, Republic of Korea Facsimile No. 82-42-472-7140	13 SEPTEMBER 2010 (13.09.2010)	JO Soo Ik Telephone No.82-42-481-5893

Form PCT/ISA/237 (cover sheet) (July 2009)

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

International application No.
PCT/US2010/022807

Box No. I Basis of this opinion

1. With regard to the **language**, this opinion has been established on the basis of :

   ☒ the international application in the language in which it was filed

   ☐ a translation of the international application into _____ , which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b))

2. ☐ This opinion has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43*bis*.1(a))

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, this opinion has been established on the basis of:

   a. a sequence listing filed or furnished
      ☐ on paper
      ☐ in electronic form

   b. time of filing or furnishing
      ☐ contained in the international application as filed.
      ☐ filed together with the international application in electronic form
      ☐ furnished subsequently to this Authority for the purposes of search.

4 ☐ In addition, in the case that more than one version or copy of a sequence listing has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.

5. Additional comments:

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

International application No.
PCT/US2010/022807

Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Claims	NONE	YES
	Claims	1-35	NO
Inventive step (IS)	Claims	NONE	YES
	Claims	1-35	NO
Industrial applicability (IA)	Claims	1-35	YES
	Claims	NONE	NO

2. Citations and explanations :

Reference is made to the following documents:

D1: US 2009-0062119 A1 (OLSON EDWIN S. et al.) 05 March 2009
D2: US 2006-0204418 A1 (CHIEN-CHUNG CHAO et al.) 14 September 2006
D3: US 2007-0234902 A1 (DAVID FAIR et al.) 11 October 2007

1. Novelty & Inventive Step

Claims [1-35] relate to a method for separating mercury from mercury containg gas.

D1 discloses methods and materials for the removal of pollutants from flue gas or product gas from a gasification system. In particular, mercury is removed from gas streams generated during the burning or gasification of fossil fuels by highly reactive regenerable sorbents.(See the abstract, fig. 1~10, Paragraph [10] ~ Paragraph [134], whole claims)

D2 discloses a catalytic adsorbents for use in the removal of mercury from flue gas streams and methods of manufacturing such catalytic adsorbents.(See the abstract, Paragraph [33] ~ Paragraph [101])

D3 discloses a method for removing mercury from a flue gas stream of the present invention is comprised of first treating a sorbent with a metal halide, and then contacting a sufficient amount of the sorbent with a gas stream for a sufficient amount of time to bind with a desired amount of the mercury in said gas stream.(See the whole document).

1.1 Claims [1-35]

1.1.1 Concerning Claim [1]

Document D1 which is considered to represent the most relevant state of the art, disclose a method of using a sorbent, a halogen promoter, a reaction process for promted halogen sorbent, a separation process.

As all of the features of claim [1] are disclosed in D1 this claim is anticipated by D1. Therefore, claim [1] lacks novelty under PCT Article 33(2).

1.1.2 Concerning Claims [2-11]

The additional feature of claims [2-11] dependent on claim [1] such as a injection and introduction of sorbent, promoter, a rate control are disclosed in D1, this claim is anticipated by D1. Therefore, claims [2-11] lack novelty under PCT Article 33(2).

Supplemental Box

In case the space in any of the preceding boxes is not sufficient.
Continuation of :

Box V

1.1.3 Concerning Claims [12-22]

The additional feature of claims [12-22] dependent on claim [1] such as a activated carbon sorbent, non carbon based sorbents, lewis base, acid group, alkaline, inorganic, sobents are disclosed in D1, this claim is anticipated by D1. Therefore, claims [12-22] lack novelty under PCT Article 33(2).

1.1.4 Concerning Claims [23-28]

The additional feature of claims [23-28] dependent on claim [1] such as carbon and non carbon promoters, a reaction process with noncarbon lewis base, acid group, alkaline, inorganic, promoted sorbent are disclosed in D1, this claim is anticipated by D1. Therefore, claims [23-28] lack novelty under PCT Article 33(2).

1.1.5 Concerning Claims [29-32]

The additional feature of claims [29-32] dependent on claim [1] such as vacuum, chmical, alkaline pretreatment process and alkaline introduction are disclosed in D1, this claim is anticipated by D1. Therefore, claims [29-32] lack novelty under PCT Article 33(2).

1.1.6 Concerning Claims [33-35]

The additional feature of claims [33-35] dependent on claim [1] such as a promoted carbon sorbent and mercury level control are disclosed in D1, this claim is anticipated by D1. Therefore, claims [33-35] lack novelty under PCT Article 33(2).

2. Industrial Applicability

The subject matter of claims [1-35] are considered to be industrially applicable under PCT Article 33(4).

PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference	FOR FURTHER ACTION	see Form PCT/ISA/220 as well as, where applicable, item 5 below.
16291.3WO01		
International application No.	International filing date *(day/month/year)*	(Earliest) Priority Date *(day/month/year)*
PCT/US2010/022807	**02 FEBRUARY 2010 (02.02.2010)**	23 APRIL 2009 (23.04.2009)

Applicant

ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION et al

This International search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 3 sheets.

☐ It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**
   a. With regard to the **language**, the international search was carried out on the basis of :
      ☒ the international application in the language in which it was filed
      ☐ a translation of the international application into _____, which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b))
   b. ☐ This international search report has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43.6*bis*(a))
   c. ☐ With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, see Box No I.
2. ☐ **Certain claims were found unsearchable** (See Box No. II)
3. ☐ **Unity of invention is lacking** (See Box No. III)
4. With regard to the **title,**
   ☒ the text is approved as submitted by the applicant.
   ☐ the text has been established by this Authority to read as follows:
5. With regard to the **abstract,**
   ☒ the text is approved as submitted by the applicant.
   ☐ the text has been established, according to Rule 38.2, by this Authority as it appears in Box No. IV. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.
6. With regard to the drawings,
   a. the figure of the **drawings** to be published with the abstract is Figure No. 3
      ☐ as suggested by the applicant
      ☒ as selected by this Authority, because the applicant failed to suggest a figure.
      ☐ as selected by this Authority, because this figure better characterizes the invention.
   b. ☐ none of the figure is to be published with the abstract.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US2010/022807

A. CLASSIFICATION OF SUBJECT MATTER

B01D 53/02(2006.01)i, B01D 53/64(2006.01)i, F23J 15/00(2006.01)i

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)

B01D 53/02; C01B 31/08; B01J 8/00; B01J 20/00

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Korean utility models and applications for utility models
Japanese utility models and applications for utility models

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

eKOMPASS(KIPO internal) & Keywords: sorbent, mercury, flue gas, halogen promoter

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2009-0062119 A1 (OLSON EDWIN S. et al.) 05 March 2009 See the abstract, fig. 1-10, Paragraph [10] - Paragraph [134], whole claims	1-35
A	US 2006-0204418 A1 (CHIEN-CHUNG CHAO et al.) 14 September 2006 See the abstract, Paragraph [33] - Paragraph [101]	1-35
A	US 2007-0234902 A1 (DAVID FAIR et al.) 11 October 2007 See the whole document	1-35

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" earlier application or patent but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of 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
13 SEPTEMBER 2010 (13.09.2010)	**13 SEPTEMBER 2010 (13.09.2010)**

Name and mailing address of the ISA/KR
Korean Intellectual Property Office
Government Complex-Daejeon, 139 Seonsa-ro, Seo-gu, Daejeon 302-701, Republic of Korea
Facsimile No. 82-42-472-7140

Authorized officer
JO Soo Ik
Telephone No. 82-42-481-5893

Form PCT/ISA/210 (second sheet) (July 2009)

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 2009-0062119 A1	05.03.2009	None	
US 2006-0204418 A1	14.09.2006	CA 2600876-A1	21.09.2006
		CN 101175692 A0	07.05.2008
		EP 1866245 A2	19.12.2007
		KR 10-2007-0113287 A	28.11.2007
		MX 2007011233 A	06.11.2007
		US 2006-0205592 A1	14.09.2006
		WO 2006-099291 A2	21.09.2006
		WO 2006-099291 A3	21.09.2006
US 2007-0234902 A1	11.10.2007	WO 2007-112248 A2	04.10.2007
		WO 2007-112248 A3	13.12.2007
		WO 2007-112248 A3	04.10.2007

PATENT COOPERATION TREATY

From the
INTERNATIONAL SEARCHING AUTHORITY

To:
KOWALCHYK KATHERINE M

MERCHANT & GOULD P.C. P.O. BOX 2903
MINNEAPOLIS MN 55402-0903 USA

PCT

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

(PCT Rule 43bis.1)

Date of mailing *(day/month/year)*	**13 SEPTEMBER 2010 (13.09.2010)**
Applicant's or agent's file reference 16291.3WO11	**FOR FURTHER ACTION** See paragraph 2 below

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/US2010/022807	**02 FEBRUARY 2010 (02.02.2010)**	23 APRIL 2009 (23.04.2009)

International Patent Classification (IPC) or both national classification and IPC

B01D 53/02(2006.01)i, B01D 53/64(2006.01)i, F23J 15/00(2006.01)i

Applicant

ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION et al

1. This opinion contains indications relating to the following items:

- [x] Box No. I Basis of the opinion
- [ ] Box No. II Priority
- [ ] Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- [ ] Box No. IV Lack of unity of invention
- [x] Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- [ ] Box No. VI Certain documents cited
- [ ] Box No. VII Certain defects in the international application
- [ ] Box No. VIII Certain observations on the international application

2. **FURTHER ACTION**

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If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of 3 months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.
For further options, see Form PCT/ISA/220.

3. For further details, see notes to Form PCT/ISA/220.

Name and mailing address of the ISA/KR	Date of completion of this opinion	Authorized officer
Korean Intellectual Property Office Government Complex-Daejeon, 139 Seonsa-ro, Seo-gu, Daejeon 302-701, Republic of Korea Facsimile No. 82-42-472-7140	13 SEPTEMBER 2010 (13.09.2010)	JO Soo Ik Telephone No.82-42-481-5893

Form PCT/ISA/237 (cover sheet) (July 2009)

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

International application No.
PCT/US2010/022807

Box No. I Basis of this opinion

1. With regard to the **language**, this opinion has been established on the basis of :

   ☒ the international application in the language in which it was filed

   ☐ a translation of the international application into _____ , which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b))

2. ☐ This opinion has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43*bis*.1(a))

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, this opinion has been established on the basis of:

   a. a sequence listing filed or furnished
      ☐ on paper
      ☐ in electronic form

   b. time of filing or furnishing
      ☐ contained in the international application as filed.
      ☐ filed together with the international application in electronic form
      ☐ furnished subsequently to this Authority for the purposes of search.

4 ☐ In addition, in the case that more than one version or copy of a sequence listing has been filed or furnished, the required statements that the information in the subsequent or additioanl copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.

5. Additional comments:

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

International application No.
PCT/US2010/022807

Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Claims	NONE	YES
	Claims	1-35	NO
Inventive step (IS)	Claims	NONE	YES
	Claims	1-35	NO
Industrial applicability (IA)	Claims	1-35	YES
	Claims	NONE	NO

2. Citations and explanations :

Reference is made to the following documents:

D1: US 2009-0062119 A1 (OLSON EDWIN S. et al.) 05 March 2009
D2: US 2006-0204418 A1 (CHIEN-CHUNG CHAO et al.) 14 September 2006
D3: US 2007-0234902 A1 (DAVID FAIR et al.) 11 October 2007

1. Novelty & Inventive Step

Claims [1-35] relate to a method for separating mercury from mercury containg gas.

D1 discloses methods and materials for the removal of pollutants from flue gas or product gas from a gasification system. In particular, mercury is removed from gas streams generated during the burning or gasification of fossil fuels by highly reactive regenerable sorbents.(See the abstract, fig. 1~10, Paragraph [10] ~ Paragraph [134], whole claims)

D2 discloses a catalytic adsorbents for use in the removal of mercury from flue gas streams and methods of manufacturing such catalytic adsorbents.(See the abstract, Paragraph [33] ~ Paragraph [101])

D3 discloses a method for removing mercury from a flue gas stream of the present invention is comprised of first treating a sorbent with a metal halide, and then contacting a sufficient amount of the sorbent with a gas stream for a sufficient amount of time to bind with a desired amount of the mercury in said gas stream.(See the whole document).

1.1 Claims [1-35]

1.1.1 Concerning Claim [1]

Document D1 which is considered to represent the most relevant state of the art, disclose a method of using a sorbent, a halogen promoter, a reaction process for promted halogen sorbent, a separation process.

As all of the features of claim [1] are disclosed in D1 this claim is anticipated by D1. Therefore, claim [1] lacks novelty under PCT Article 33(2).

1.1.2 Concerning Claims [2-11]

The additional feature of claims [2-11] dependent on claim [1] such as a injection and introduction of sorbent, promoter, a rate control are disclosed in D1, this claim is anticipated by D1. Therefore, claims [2-11] lack novelty under PCT Article 33(2).

Supplemental Box

In case the space in any of the preceding boxes is not sufficient.
Continuation of :

Box V

1.1.3 Concerning Claims [12-22]

The additional feature of claims [12-22] dependent on claim [1] such as a activated carbon sorbent, non carbon based sorbents, lewis base, acid group, alkaline, inorganic, sobents are disclosed in D1, this claim is anticipated by D1. Therefore, claims [12-22] lack novelty under PCT Article 33(2).

1.1.4 Concerning Claims [23-28]

The additional feature of claims [23-28] dependent on claim [1] such as carbon and non carbon promoters, a reaction process with noncarbon lewis base, acid group, alkaline, inorganic, promoted sorbent are disclosed in D1, this claim is anticipated by D1. Therefore, claims [23-28] lack novelty under PCT Article 33(2).

1.1.5 Concerning Claims [29-32]

The additional feature of claims [29-32] dependent on claim [1] such as vacuum, chmical, alkaline pretreatment process and alkaline introduction are disclosed in D1, this claim is anticipated by D1. Therefore, claims [29-32] lack novelty under PCT Article 33(2).

1.1.6 Concerning Claims [33-35]

The additional feature of claims [33-35] dependent on claim [1] such as a promoted carbon sorbent and mercury level control are disclosed in D1, this claim is anticipated by D1. Therefore, claims [33-35] lack novelty under PCT Article 33(2).

2. Industrial Applicability

The subject matter of claims [1-35] are considered to be industrially applicable under PCT Article 33(4).

PATENT COOPERATION TREATY

Schwegman Lundberg & Woessner P.A.
JUL 24 2014
PCT RECEIVED

From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

SCHWEGMAN, LUNDBERG & WOESSNER, P.A.
P.O. Box 2938
Minneapolis, MN 55402
ETATS-UNIS D'AMERIQUE

NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(PCT Rule 71.1)

Date of mailing *(day/month/year)*	17.07.2014
Applicant's or agent's file reference	IMPORTANT NOTIFICATION
3338016WO1	

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/US2013/036964	17.04.2013	23.04.2012

Applicant
Energy & Environmental Research Center Foundation

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary report on patentability and its annexes, if any, established on the international application.

2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.

3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

4. **REMINDER**

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary report on patentability. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

The applicant's attention is drawn to Article 33(5), which provides that the criteria of novelty, inventive step and industrial applicability described in Article 33(2) to (4) merely serve the purposes of international preliminary examination and that "any Contracting State may apply additional or different criteria for the purposes of deciding whether, in that State, the claimed inventions is patentable or not" (see also Article 27(5)). Such additional criteria may relate, for example, to exemptions from patentability, requirements for enabling disclosure, clarity and support for the claims.

Name and mailing address of the international preliminary examining authority:

European Patent Office P.B. 5818 Patentlaan 2
NL-2280 HV Rijswijk - Pays Bas
Tel. +31 70 340 - 2040
Fax: +31 70 340 - 3016

Authorized Officer

Bergström, Carina

Tel. +31 70 340-2898

Form PCT/IPEA/416 (January 2004)

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference 3338016WO1	**FOR FURTHER ACTION**	See Form PCT/IPEA/416
International application No. PCT/US2013/036964	International filing date *(day/month/year)* 17.04.2013	Priority date *(day/month/year)* 23.04.2012

International Patent Classification (IPC) or national classification and IPC
INV. B01J20/12

Applicant
Energy & Environmental Research Center Foundation

1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 10 sheets, including this cover sheet.
3. This report is also accompanied by ANNEXES, comprising:
   a. ☒ *(sent to the applicant and to the International Bureau)* a total of 13 sheets, as follows:
      - ☐ sheets of the description, claims and/or drawings which have been amended and/or sheets containing rectifications authorized by this Authority, unless those sheets were superseded or cancelled, and any accompanying letters (see Rules 46.5, 66.8, 70.16, 91.2, and Section 607 of the Administrative Instructions).
      - ☐ sheets containing rectifications, where the decision was made by this Authority not to take them into account because they were not authorized by or notified to this Authority at the time when this Authority began to draw up this report, and any accompanying letters (Rules 66.4bis, 70.2(e), 70.16 and 91.2).
      - ☒ superseded sheets and any accompanying letters, where this Authority either considers that the superseding sheets contain an amendment that goes beyond the disclosure in the international application as filed, or the superseding sheets were not accompanied by a letter indicating the basis for the amendments in the application as filed, as indicated in item 4 of Box No. I and the Supplemental Box (see Rule 70.16(b)).

   b. ☐ *(sent to the International Bureau only)* a total of (indicate type and number of electronic carrier(s)) , containing a sequence listing, in electronic form only, as indicated in the Supplemental Box Relating to Sequence Listing (see paragraph 3*bis* of Annex C of the Administrative Instructions).

4. This report contains indications relating to the following items:
   - ☒ Box No. I Basis of the report
   - ☐ Box No. II Priority
   - ☒ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - ☐ Box No. IV Lack of unity of invention
   - ☒ Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
   - ☐ Box No. VI Certain documents cited
   - ☒ Box No. VII Certain defects in the international application
   - ☒ Box No. VIII Certain observations on the international application

Date of submission of the demand	Date of completion of this report
28.02.2014	17.07.2014

Name and mailing address of the international preliminary examining authority:
European Patent Office P.B. 5818 Patentlaan 2
NL-2280 HV Rijswijk - Pays Bas
Tel. +31 70 340 - 2040
Fax: +31 70 340 - 3016

Authorized officer
Klemps, Christian
Telephone No. +31 70 340-8036

Box No. I Basis of the report

1. With regard to the **language**, this report is based on

☒ the international application in the language in which it was filed

☐ a translation of the international application into , which is the language of a translation furnished for the purposes of:

- ☐ international search (under Rules 12.3(a) and 23.1(b))
- ☐ publication of the international application (under Rule 12.4(a))
- ☐ international preliminary examination (under Rules 55.2(a) and/or 55.3(a) and (b))

2. With regard to the **elements*** of the international application, this report is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report)*:

Description, Pages

1-49 as originally filed

Claims, Numbers

1-42 filed with the demand for preliminary international examination

Drawings, Sheets

1/6-6/6 as originally filed

☐ a sequence listing - see Supplemental Box Relating to Sequence Listing.

3. ☐ The amendments have resulted in the cancellation of:

- ☐ the description, pages
- ☐ the claims, Nos.
- ☐ the drawings, sheets/figs
- ☐ the sequence listing *(specify)*:
- ☐ any table(s) related to sequence listing *(specify)*:

4. ☒ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since either they are considered to go beyond the disclosure as filed, or they were not accompanied by a letter indicating the basis for the amendments in the application as filed, as indicated in the Supplemental Box (Rules 70.2(c) and (c-bis)):

- ☐ the description, pages
- ☒ the claims, Nos. 1,38,41
- ☐ the drawings, sheets/figs
- ☐ the sequence listing *(specify)*:

5. ☐ This report has been established:

- ☐ taking into account the rectification of an obvious mistake authorized by or notified to this Authority under Rule 91 (Rules 66.1(d-bis) and 70.2(e)).
- ☐ without taking into account the rectification of an obvious mistake authorized by or notified to this Authority under Rule 91(Rules 66.4bis and 70.2(e)).

6. ☒ With regard to top-up searches (Rules 66.1 ter and 70.2(f)):
   - ☒ A top-up search was carried out by this Authority on 08.04.2014 (all discovered documents are listed in the Supplemental Box Relating to Top-up Search).
   - ☐ No top-up search was carried out by this Authority.

7. ☐ Supplementary international search report(s) from Authority(ies) has/have been received and taken into account in establishing this report (Rule 45bis.8(b) and (c)).

Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1. The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:

   ☐ the entire international application,

   ☒ claims Nos. 6-43, 46, 47, 49

   because:

   ☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination *(specify)*:

   ☐ the description, claims or drawings *(indicate particular elements below)* or said claims Nos. are so unclear that no meaningful opinion could be formed *(specify)*:

   ☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed *(specify)*.

   ☒ no international search report has been established for the said claims Nos. 6-43, 46, 47, 49

   ☐ a meaningful opinion could not be formed without the sequence listing; the applicant did not, within the prescribed time limit:

      ☐ furnish a sequence listing on paper complying with the standard provided for in Annex C of the Administrative Instructions, and such listing was not available to the International Preliminary Examining Authority in a form and manner acceptable to it.

      ☐ furnish a sequence listing in electronic form complying with the standard provided for in Annex C of the Administrative Instructions, and such listing was not available to the International Preliminary Examining Authority in a form and manner acceptable to it.

      ☐ pay the required late furnishing fee for the furnishing of a sequence listing in response to an invitation under Rules 13*ter*.1(a) or (b) and 13*ter*.2.

   ☒ See separate sheet for further details

Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	
	No:	Claims	1-5, 44, 46, 48
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1-5, 44, 46, 48
Industrial applicability (IA)	Yes:	Claims	1-5, 44, 46, 48
	No:	Claims	

2. Citations and explanations (Rule 70.7):

see separate sheet

Box No. VII Certain defects in the international application

The following defects in the form or contents of the international application have been noted:

see separate sheet

Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

Re Item I

Basis of the report

1 The International Preliminary Examining Authority considers that the following amendments to the claims go beyond the disclosure in the international application as filed:

In present claims 1 and 38, the "substrate material" is defined as "comprising at least one of diatomaceous earth, a clay, a zeolite, and a mineral". In paragraph [0061] of the description of the application as filed, however, only porous minerals are disclosed. No basis in the application as filed can be found for *any* mineral as "substrate material".

Further, in present claims 1 and 38, the "heat treatment" is defined as "comprising about 500°C to about 1200°C". In paragraph [0065] of the description of the application as filed, however, only each of the temperatures 500°C and 1200°C, respectively, are disclosed in an exemplified fashion. No basis in the application as filed can be found for a *temperature range from 500°C to 1200°C* of said "heat treatment".

Further, in present claim 1, the "promotion comprising chemical reaction with at least one of a halogen and a halide promotor" is defined as taking place "after the heat treatment". However, in none of the paragraphs [0058], [0061], [0065] or [0066] - [0074] mentioned in the applicant's letter as basis for this amendment said "promotion comprising chemical reaction with at least one of a halogen and a halide promotor" is specifically disclosed as taking place *after the heat treatment*.

The present opinion therefore has been established as if such amendments had not been made (Rule 70.2(c) PCT).

2 Claim 41 has been newly introduced. Since in the accompanying letter, the basis in the application as filed for this amendment was not indicated, this report has been established as if this amendment had not been made (Rule 70.2(c-bis).

3 Consequently, the present report has been established on the application documents as originally filed.

Re Item III

Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

4 No opinion was established with regard to novelty, inventive step and industrial applicability on the subject-matter of claims 4-37, 40 and 42 since their subject-matter relates to inventions for which no international search report has been established (Rule 66.2(e) PCT).

Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

Reference is made to the following documents:

D1 US 2011/168018 A1 (MOHAMADALIZADEH ALI [IR] ET AL) 14 July 2011 (2011-07-14)

D2 US 2009/136401 A1 (YANG XIAOLIN DAVID [US] ET AL) 28 May 2009 (2009-05-28)

D3 US 2004/076570 A1 (JIA CHARLES Q [CA]) 22 April 2004 (2004-04-22)

D4 A. Bakandritsos ET AL: "High Surface Area Montmorillonite-Carbon Composites and Derived Carbons", Chemistry of materials, vol. 16, no. 8, 16 March 2004 (2004-03-16), pages 1551-1559, XP055072551, ISSN: 0897-4756, DOI: 10.1021/cm0350030

D5 US 2008/090951 A1 (MAO DONGSHENG [US] ET AL) 17 April 2008 (2008-04-17)

5 For the reasons mentioned under item I (see above), the present reasoned statement with regard to novelty, inventive step or industrial applicability has been established on the first (group of) inventions mentioned in the claims as originally filed. The claim numbering relates to those said claims as originally filed:

6 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 1-5, 44, 45 and 48 is not new in the sense of Article 33(2) PCT:

7 **D1** discloses a method for separating hydrogen sulfide (a pollutant) from a gas comprising methane, helium and hydrogen sulfide, the method comprising

providing a carbon nanocomposite sorbent of multiwall carbon nanotubes (MWCNT) with average diameters of from about 2 nm to about 5 nm and mesoporous crystalline material (MCM)-41;

contacting the said carbon nanocomposite sorbent with a flow of the gas containing 5500 ppm of hydrogen sulfide;

passing the gas flow to an outlet to give a separated gas containing 600 ppm of hydrogen sulfide (paragraph [0029]; paragraph [0062]; example 2).

D1 further discloses a method of making a carbon nanocomposite sorbent and the carbon nanocomposite sorbent hence made, the method comprising

providing MWCNT as carbon precursor;

providing MCM-41 as substrate material;

mixing MWCNTs and MCM-41 and thereby contacting them;

heating the mixture for 3 hours at 110°C (paragraph [0029]; paragraph [0062]; example 2).

D1 hence discloses the subject-matter of claims 1, 2, 44, 45 and 48.

8 **D2** discloses discloses a method for separating mercury from a flue gas comprising flue gas, fly ash and mercury, the method comprising

providing as-mined kaolin containing about 2% naturally intercalated carbon (Kaolin-1), which is considered as a carbon nanocomposite sorbent;

promoting the said carbon nanocomposite sorbent by impregnating it with a sodium bromide (NaBr) solution in water;

contacting the NaBr-promoted carbon nanocomposite sorbent with a flow of the gas containing mercury for one second by injecting the carbon nanocomposite sorbent into the gas in a drop tube reactor;

separating the NaBr-promoted carbon nanocomposite sorbent from the gas for analysis of mercury content (paragraph [0044] - paragraph [0047]; examples 8, 16; tables 1, 2; paragraph [0048] - paragraph [0050]; table 3).

D2 further discloses as-mined kaolin containing about 2% naturally intercalated carbon (Kaolin-1), which is considered as a carbon nanocomposite sorbent (paragraph [0044] - paragraph [0047]; examples 8; table 1).

D2 hence discloses the subject-matter of claims 1-5, 45 and 48.

9 D3 discloses a method of separating mercury from a desiccator atmosphere that comprises mercury, the method comprising

providing treated coke (ECOcarbon), which is considered a carbon nanocomposite comprising agglomerated carbon structures of about 30 nm;

contacting the said carbon nanocomposite sorbent with a desiccator atmosphere that comprises mercury;

separating the said carbon nanocomposite sorbent comprising mercury from the desiccator atmosphere for analysis of mercury content (paragraph [0084] - paragraph [0086]; example 1; paragraph [0095] - paragraph [0099]; example 5).

D3 further discloses a method of making a carbon nanocomposite sorbent and the carbon nanocomposite sorbent hence made, the method comprising

providing fluid coke as carbon precursor;

providing sulfur dioxide (SO_2) as substrate material;

contacting fluid coke and SO_2 for 10 seconds;

heating the mixture at 800°C (paragraph [0087] - paragraph [0092]; example 2).

D3 hence discloses the subject-matter of claims 1-3, 44, 45 and 48.

10 D4 discloses a method of making a carbon nanocomposite sorbent and the carbon nanocomposite sorbent hence made, the method comprising

providing sucrose as carbon precursor;

providing montmorillonite as substrate material;

contacting contacting sucrose and montmorillonite;

pyrolyzing and hence heating the mixture for one hour at 600°C (Section: "Sample Preparation";page 1552, right-hand column; figure 1).

D4 hence discloses the subject-matter of claims 44, 45 and 48.

11 D5 discloses a method of making a carbon nanocomposite suitable as a sorbent and the carbon nanocomposite hence made, the method comprising

providing carbon nanotubes (CNTs) as carbon precursors,

providing epoxy as substrate material,

curing and hence heating the material for two hours at 160°C .

D5 hence discloses the subject-matter of claims 44, 45 and 48 (paragraph [0015]; figure 1).

Re Item VII

Certain defects in the international application

12 Contrary to the requirements of Rule 5.1(a)(ii) PCT, the relevant background art documents mentioned in paragraphs [0005] to [0007] of the description are not cited in a way that allows their retrieval.

Re Item VIII

Certain observations on the international application

13 For the reasons mentioned under Item I (see above), the present observations have been made with regard to the first (group of) inventions mentioned in the claims as originally filed. The claim numbering relates to those said claims as originally filed:

14 The application does not meet the requirements of Article 6 PCT for the following reasons:

15 The term "carbon nanocomposite" use in the claims is unclear, because it cannot be determined without ambiguity which further technical feature is meant to be attributed to the carbon composite by defining it as a "nano"composite. The term "nano" merely indicates a prefix in the SI and other systems of units denoting a factor of 10^{-9}. For the present purposes, any carbon composite is considered to be a "carbon nanocomposite". Insofar as

the description of the application contains a clear and unambiguous definition of the tem "carbon nanocomposite", said definition would have to be included in the claims to render the subject-matter of said claim clear.

16 The term pollutant used in claim 2 is vague and unclear and leaves the reader in doubt as to the meaning of the technical feature to which it refers. Specifically, whether a compound or material is considered to be polluting or not, depends on the specific circumstances in which the compound or material is found. Since theses specific circumstances are not further defined in the claim, the said claim is unclear. Insofar as the description of the application contains a clear and unambiguous definition of the tem "pollutant", said definition would have to be included in the claims to render the subject-matter of said claim clear.

17 Claim 4 is not supported by the description as required by Article 6 PCT, as its scope is broader than justified by the description and drawings. Specific support for "promoters" can only be found for "halogen and halide" promoters (see e.g. paragraph [0009] of the description), not for any "promoter".

Claim 48 tries to define the claimed carbon nanocomposite sorbent further in that it is "in contact with mercury or oxidized mercury". This wording introduces an ambiguity as to the scope of said claim 48. The expression "in contact" seeks to define a relationship between the entities "carbon nanocomposite sorbent" and "mercury or oxidized mercury", while it cannot be determined whether one of the two or both entities fall under the scope of the claim. The subject-matter of claim 48 therefore is unclear. For the present purposes of search and examination, claim 48 is considered to be solely directed to the "carbon nanocomposite sorbent" of claim 45, upon which it depends.

PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference	**FOR FURTHER ACTION**	see Form PCT/ISA/220 as well as, where applicable, item 5 below.
3338016WO1		
International application No.	International filing date *(day/month/year)*	(Earliest) Priority Date *(day/month/year)*
PCT/US2013/036964	17 April 2013 (17-04-2013)	23 April 2012 (23-04-2012)
Applicant		
ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION		

This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 7 sheets.

☐ It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**

a. With regard to the **language**, the international search was carried out on the basis of:

☒ the international application in the language in which it was filed

☐ a translation of the international application into _____, which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b))

b. ☐ This international search report has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43.6*bis*(a)).

c. ☐ With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, see Box No. I.

2. ☐ **Certain claims were found unsearchable** (See Box No. II)

3. ☒ **Unity of invention is lacking** (see Box No III)

4. With regard to the **title**,

☒ the text is approved as submitted by the applicant

☐ the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

☒ the text is approved as submitted by the applicant

☐ the text has been established, according to Rule 38.2, by this Authority as it appears in Box No. IV. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority

6. With regard to the **drawings**,

a. the figure of the **drawings** to be published with the abstract is Figure No. 1

☒ as suggested by the applicant

☐ as selected by this Authority, because the applicant failed to suggest a figure

☐ as selected by this Authority, because this figure better characterizes the invention

b. ☐ none of the figures is to be published with the abstract

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US2013/036964

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

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

see additional sheet

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying an additional fees, this Authority did not invite payment of additional fees.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

1-5, 44, 45, 48

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.

☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.

☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2013/036964

A. CLASSIFICATION OF SUBJECT MATTER
INV. B01J20/12 B01D53/02 B01J20/30 B01J20/32 B01D53/64
B01J20/20
ADD.
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)
B01J B01D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
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.
X	US 2011/168018 A1 (MOHAMADALIZADEH ALI [IR] ET AL) 14 July 2011 (2011-07-14) paragraph [0029] paragraph [0062]; example 2 -----	1,2,44, 45,48
X	US 2009/136401 A1 (YANG XIAOLIN DAVID [US] ET AL) 28 May 2009 (2009-05-28) paragraph [0044] - paragraph [0047]; examples 8, 16; tables 1, 2 paragraph [0048] - paragraph [0050]; table 3 ----- -/--	1-5,45, 48

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

[X] See patent family annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
25 July 2013	29/11/2013

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, Fax: (+31-70) 340-3016	Klemps, Christian

Form PCT/ISA/210 (second sheet) (April 2005)

PACIRA EXHIBIT 1023
Page 528

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2004/076570 A1 (JIA CHARLES Q [CA]) 22 April 2004 (2004-04-22) paragraph [0084] - paragraph [0086]; example 1 paragraph [0087] - paragraph [0092]; example 2 paragraph [0095] - paragraph [0099]; example 5	1-3,44, 45,48
X	A. BAKANDRITSOS ET AL: "High Surface Area Montmorillonite-Carbon Composites and Derived Carbons", CHEMISTRY OF MATERIALS, vol. 16, no. 8, 16 March 2004 (2004-03-16) , pages 1551-1559, XP055072551, ISSN: 0897-4756, DOI: 10.1021/cm0350030 Section: "Sample Preparation"; page 1552, right-hand column; figure 1	44,45,48
X	US 2008/090951 A1 (MAO DONGSHENG [US] ET AL) 17 April 2008 (2008-04-17) paragraph [0015]; figure 1	44,45,48

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/US2013/036964

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 2011168018 A1	14-07-2011	NONE	
US 2009136401 A1	28-05-2009	US 2009136401 A1	28-05-2009
		US 2012292564 A1	22-11-2012
US 2004076570 A1	22-04-2004	AU 2003278010 A1	04-05-2004
		CA 2408858 A1	18-04-2004
		US 2004076570 A1	22-04-2004
		WO 2004035176 A1	29-04-2004
US 2008090951 A1	17-04-2008	NONE	

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. claims: 1-5, 44, 45, 48

related to a method for separating a material from a gas that comprises the material, the method comprising providing or obtaining a carbon nanocomposite sorbent, contacting at least some of the material with the sorbent, to form a material-sorbent composition, and separating at least some of the material-sorbent composition from the material-containing gas, to give a separated gas; further to a method of making a such carbon nanocomposite sorbent; further to a such carbon nanocomposite

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2. claims: 6-42, 46, 49

related to a method for reducing the pollutant content of a pollutant-containing gas, the method comprising providing or obtaining a carbon nanocomposite sorbent material, providing or obtaining a halogen or halide promotor, promoting at least a portion of the sorbent material by chemically reacting the portion of the sorbent material with the halogen or halide promoter to form a promoted sorbent, contacting at least part of the promoted sorbent with the pollutant in the pollutant-containing gas, to form a pollutant-sorbent composition, and separating particulates from the pollutant-containing gas, the particulates comprising at least some of the pollutant-sorbent composition, to give a cleaned gas; further to a promoted corbon nanocomposite sorbent made by reacting a portion of the sorbent material with a halogen or halide promoter to form the promoted sorbent

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3. claims: 43, 47

related to a method for reducing the mercury content of a mercury-containing gas, the method comprising providing or obtaining a carbon nanocomposite sorbent, the carbon nanocomposite sorbent made by steps comprising providing or obtaining a carbon precursor, providing or obtaining a substrate material, contacting the carbon precursor and the substrate material, to provide a nanocomposite starting material, and heating the nanocomposite starting material, to provide the carbon nanocomposite sorbent, providing or obtaining a halogen or halide promoter, promoting at least a portion of the sorbent material by chemically reacting the portion of the sorbent material with the halogen or halide promoter to form a promoted sorbent, contacting at least part of the promoted sorbent with the mercury, to form a mercury-sorbent composition, and separating at least some of the mercury-sorbent composition from the mercury-containing gas; further to a mercury-sorbent composition made by

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

contacting a carbon nanocomposite sorbent with a mercury-containing gas

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PATENT COOPERATION TREATY

From the INTERNATIONAL SEARCHING AUTHORITY

PCT

To:
Madden, Robert B. Reg. No. 57,521
SCHWEGMAN, LUNDBERG & WOESSNER
P.O. Box 2938
Minneapolis, MN 55402-0938
ETATS-UNIS D'AMERIQUE

INVITATION TO PAY ADDITIONAL FEES AND, WHERE APPLICABLE, PROTEST FEE

(PCT Article 17(3)(a) and Rule 40.1 and 40.2(e))

Applicant's or agent's file reference 3338016WO1	Date of mailing *(day/month/year)* 2 August 2013 (02-08-2013)
International application No. PCT/US2013/036964	**PAYMENT DUE** within **ONE MONTH** from the above date of mailing
Applicant ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION	International filing date *(day/month/year)* 17 April 2013 (17-04-2013)

1. This International Searching Authority

(i) considers that there are 3 *(number of)* inventions claimed in the international application covered by the claims indicated on an extra sheet:

(ii) therefore considers that **the international application does not comply with the requirements of unity of invention** (Rules 13.1, 13.2 and 13.3) for the reasons indicated on an extra sheet:

(iii) [X] has carried out a partial international search (see Annex) [ ] will establish the international search report on those parts of the international application which relate to the invention first mentioned in claims Nos.:

see extra sheet

(iv) will establish the international search report on the other parts of the international application only if, and to the extent to which, additional fees are paid.

2. Consequently, the applicant is hereby **invited to pay**, within the time limit indicated above, the amount indicated below:

EUR 1.875,00	x	2	=	EUR 3.750,00
Fee per additional invention		number of additional inventions		currency/total amount of additional fees

3. The applicant is informed that, according to Rule 40.2(c), **the payment of any additional fee may be made under protest**, i.e., a reasoned statement to the effect that the international application complies with the requirement of unity of invention or that the amount of the required additional fee is excessive, where applicable, subject to the payment of a protest fee.

Where the applicant pays additional fees under protest, the applicant is hereby invited, within the time limit indicated above, to pay a protest fee (Rule 40.2(e)) in the amount of EUR 830,00 *(currency/amount)*

Where the applicant has not, within the time limit indicated above, paid the required protest fee, the protest will be considered not to have been made and the International Searching Authority will so declare.

4. [ ] Claim(s) Nos. _____ have been found to be unsearchable under Article 17(2)(b) because of defects under Article 17(2)(a) and therefore have not been included in the opinion.

Name and mailing address of the International Searching Authority	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk Tel. (+31-70) 340-2040 Fax: (+31-70) 340-3016	NAMAGEMBE, Victoria Tel: +31 (0)70 340-3963

Schwegman Lundberg & Woessner P.A.
AUG 12 2013
RECEIVED

Form PCT/ISA/206 (April 2005)
Registered Mail

PETITIONER'S EXHIBIT 1023

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. claims: 1-5, 44, 45, 48

related to a method for separating a material from a gas that comprises the material, the method comprising providing or obtaining a carbon nanocomposite sorbent, contacting at least some of the material with the sorbent, to form a material-sorbent composition, and separating at least some of the material-sorbent composition from the material-containing gas, to give a separated gas; further to a method of making a such carbon nanocomposite sorbent; further to a such carbon nanocomposite

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2. claims: 6-42, 46, 49

related to a method for reducing the pollutant content of a pollutant-containing gas, the method comprising providing or obtaining a carbon nanocomposite sorbent material, providing or obtaining a halogen or halide promotor, promoting at least a portion of the sorbent material by chemically reacting the portion of the sorbent material with the halogen or halide promoter to form a promoted sorbent, contacting at least part of the promoted sorbent with the pollutant in the pollutant-containing gas, to form a pollutant-sorbent composition, and separating particulates from the pollutant-containing gas, the particulates comprising at least some of the pollutant-sorbent composition, to give a cleaned gas; further to a promoted corbon nanocomposite sorbent made by reacting a portion of the sorbent material with a halogen or halide promoter to form the promoted sorbent

---

3. claims: 43, 47

related to a method for reducing the mercury content of a mercury-containing gas, the method comprising providing or obtaining a carbon nanocomposite sorbent, the carbon nanocomposite sorbent made by steps comprising providing or obtaining a carbon precursor, providing or obtaining a substrate material, contacting the carbon precursor and the substrate material, to provide a nanocomposite starting material, and heating the nanocomposite starting material, to provide the carbon nanocomposite sorbent, providing or obtaining a halogen or halide promoter, promoting at least a portion of the sorbent material by chemically reacting the portion of the sorbent material with the halogen or halide promoter to form a promoted sorbent, contacting at least part of the promoted sorbent with the mercury, to form a mercury-sorbent composition, and separating at least some of the mercury-sorbent composition from the mercury-containing

gas; further to a mercury-sorbent composition made by contacting a carbon nanocomposite sorbent with a mercury-containing gas

---

The present application lacks unity of invention (Rule 13 PCT) for the following reasons:
The present application contains the following eight independent claims: claim 1 (method claim), claim 6 (method claim), claim 43 (method claim), claim 44 (method claim), claim 45 (product claim), claim 46 (product claim), claim 47 (product claim), claim 49 (product claim).
The common same or corresponding technical features among those claims are:
carbon nanocomposite sorbent; method where said carbon nanocomposite sorbent is employed with a gas; said method comprising: the step of providing or obtaining a carbon nanocomposite sorbent; the step of contacting at least part of the sorbent; the step of separating from gas.
The common same or corresponding technical feature "carbon nanocomposite" is unclear (Article 6 PCT), because it cannot be determined which further technical feature is meant to be attributed to the carbon composite by defining it as a "nano"composite. The term "nano" merely indicates a prefix in the SI and other systems of units denoting a factor of 10^-9 . For the present purposes, any carbon composite is considered to be a "carbon nanocomposite".
The document US 2011/0168018 A1 (hereinafter D1) discloses a method for separating hydrogen sulfide from a gas comprising methane, helium and hydrogen sulfide, the method comprising
providing a carbon nanocomposite sorbent of multiwall carbon nanotubes (MWCNT) with average diameters of from about 2 nm to about 5 nm and mesoporous crystalline material (MCM)-41;
contacting the said carbon nanocomposite sorbent with a flow of the gas containing 5500 ppm of hydrogen sulfide;
passing the gas flow to an outlet to give a separated gas containing 600 ppm of hydrogen sulfide (paragraph [0029]; paragraph [0062]; example 2).
Further, the document US 2009/0136401 A1 (hereinafter D2) discloses a method for separating mercury from a flue gas comprising flue gas, fly ash and mercury, the method comprising
providing as-mined kaolin containing about 2% naturally intercalated carbon (Kaolin-1), which is considered as a carbon nanocomposite sorbent;
promoting the said carbon nanocomposite sorbent by impregnating it with a sodium bromide (NaBr) solution in water;
contacting the NaBr-promoted carbon nanocomposite sorbent with a flow of the gas containing mercury for one second by injecting the carbon nanocomposite sorbent into the gas in a drop tube reactor;
separating the NaBr-promoted carbon nanocomposite sorbent from the gas for analysis of mercury content (paragraph [0044] - paragraph [0047]; examples 8, 16; tables 1, 2; paragraph [0048] - paragraph [0050]; table 3).
All common same or corresponding technical features are known from D1 and D2. These features therefore cannot serve as "special" technical features within the meaning of Rule 13.2 PCT.
Therefore, there is no common, same or corresponding special technical feature among all independent claims and thus the requirement of Rule 13.2 PCT is not met.
Amongst the independent claims 6, 43, 46, 47 and 49, the further common

same or corresponding technical features are:
promoter; providing a halogen or halide promoter; promoting at least a portion of the carbon nanocomposite sorbent material by chemically reacting a (the) portion of the sorbent material with the halogen or halide promoter; promoted sorbent.
All common same or corresponding technical features are known from D2. These features therefore cannot serve as "special" technical features within the meaning of Rule 13.2 PCT.
Therefore, there is no common, same or corresponding special technical feature among all independent claims 6, 43, 46, 47 and 49 and thus the requirement of Rule 13.2 PCT is not met.
The separate groups of inventions are hence as mentioned above.
The common problem underlying groups 1, 2 and 3 of inventions is seen to lie in the provision of a method for the removal of a (pollutant) component of a gas from said gas and the provision of a sorbent for a such removal.
The common problem is already known from both D1 (see e.g. abstract) and D2 (see e.g. abstract). No additional problem could be identified common to these three groups of inventions which could serve as the general inventive concept as required by Rule 13.1 PCT.
The application hence does not meet the requirements of unity of invention as defined in Rule 13.1 and 13.2 PCT.

Annex to Form PCT/ISA/206
COMMUNICATION RELATING TO THE RESULTS OF THE PARTIAL INTERNATIONAL SEARCH

International Application No
PCT/US2013/036964

1. The present communication is an Annex to the invitation to pay additional fees (Form PCT/ISA/206). It shows the results of the international search established on the parts of the international application which relate to the invention first mentioned in claims Nos.:
see 'Invitation to pay additional fees'
2. This communication is not the international search report which will be established according to Article 18 and Rule 43.

3. If the applicant does not pay any additional search fees, the information appearing in this communication will be considered as the result of the international search and will be included as such in the international search report.

4. If the applicant pays additional fees, the international search report will contain both the information appearing in this communication and the results of the international search on other parts of the international application for which such fees will have been paid.

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2011/168018 A1 (MOHAMADALIZADEH ALI [IR] ET AL) 14 July 2011 (2011-07-14) paragraph [0029] paragraph [0062]; example 2	1,2,44, 45,48
X	US 2009/136401 A1 (YANG XIAOLIN DAVID [US] ET AL) 28 May 2009 (2009-05-28) paragraph [0044] - paragraph [0047]; examples 8, 16; tables 1, 2 paragraph [0048] - paragraph [0050]; table 3	1-5,45, 48
X	US 2004/076570 A1 (JIA CHARLES Q [CA]) 22 April 2004 (2004-04-22) paragraph [0084] - paragraph [0086]; example 1 paragraph [0087] - paragraph [0092]; example 2 paragraph [0095] - paragraph [0099]; example 5	1-3,44, 45,48
X	A. BAKANDRITSOS ET AL: "High Surface Area Montmorillonite-Carbon Composites and Derived Carbons", CHEMISTRY OF MATERIALS, vol. 16, no. 8, 16 March 2004 (2004-03-16) , pages 1551-1559, XP055072551, ISSN: 0897-4756, DOI: 10.1021/cm0350030 Section: "Sample Preparation"; page 1552, right-hand column; figure 1	44,45,48

-/--

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

[X] 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

4

Annex to Form PCT/ISA/206
COMMUNICATION RELATING TO THE RESULTS
OF THE PARTIAL INTERNATIONAL SEARCH

International Application No
PCT/US2013/036964

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2008/090951 A1 (MAO DONGSHENG [US] ET AL) 17 April 2008 (2008-04-17) paragraph [0015]; figure 1	44,45,48

4

Patent Family Annex

Information on patent family members

International Application No
PCT/US2013/036964

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 2011168018 A1	14-07-2011	NONE	
US 2009136401 A1	28-05-2009	US 2009136401 A1	28-05-2009
		US 2012292564 A1	22-11-2012
US 2004076570 A1	22-04-2004	AU 2003278010 A1	04-05-2004
		CA 2408858 A1	18-04-2004
		US 2004076570 A1	22-04-2004
		WO 2004035176 A1	29-04-2004
US 2008090951 A1	17-04-2008	NONE	

PATENT COOPERATION TREATY

From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

SCHWEGMAN, LUNDBERG & WOESSNER, P.A.
P.O. Box 2938
Minneapolis, MN 55402
ETATS-UNIS D'AMERIQUE

PCT

WRITTEN OPINION OF THE INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

(PCT Rule 66)

Date of mailing *(day/month/year)*	15.04.2014
Applicant's or agent's file reference: 3338016WO1	**REPLY DUE** within 1 month(s) and 15 days from the above date of mailing

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/US2013/036964	17.04.2013	23.04.2012

International Patent Classification (IPC) or both national classification and IPC
INV. B01J20/12

Applicant
Energy & Environmental Research Center Foundation

1. ☒ The written opinion established by the International Searching Authority:
   ☒ is ☐ is not
   considered to be a written opinion of the International Preliminary Examining Authority.

2. This second opinion contains indications relating to the following items:
   - ☒ Box No. I Basis of the opinion
   - ☐ Box No. II Priority
   - ☒ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - ☐ Box No. IV Lack of unity of invention
   - ☒ Box No. V Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
   - ☐ Box No. VI Certain documents cited
   - ☒ Box No. VII Certain defects in the international application
   - ☒ Box No. VIII Certain observations on the international application

3. The applicant is hereby **invited to reply** to this opinion.

   When? See the time limit indicated above. The applicant may, before the expiration of that time limit, request this Authority to grant an extension, see Rule 66.2(e).
   How? By submitting a written reply, accompanied, where appropriate, by amendments, according to Rule 66.3. For the form and the language of the amendments, see Rules 55.3 and 66.8.
   Also: For the examiner's obligation to consider amendments and/or arguments, see Rule 66.4*bis*. For an informal communication with the examiner, see Rule 66.6. For an additional opportunity to submit amendments, see Rule 66.4.
   If no reply is filed, the international preliminary examination report will be established on the basis of this opinion.

4. The final date by which the international preliminary report on patentability (Chapter II of the PCT) must be established according to Rule 69.2 is: 23.06.2014

Schwegman Lundberg & Woessner P.A.
APR 29 2014

Name and mailing address of the international preliminary examining authority:	Authorized Officer
European Patent Office P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk - Pays Bas Tel. +31 70 340 - 2040 Fax: +31 70 340 - 3016	RECEIVED Klemps, Christian Telephone No. +31 70 340-8036

Form PCT/IPEA/408 (Cover Sheet) (July 2011)

Box No. I Basis of the opinion

1. With regard to the **language**, this opinion has been established on the basis of:

   ☒ the international application in the language in which it was filed

   ☐ a translation of the international application into ,
   which is the language of a translation furnished for the purposes of:
   - ☐ international search (under Rules 12.3(a) and 23.1(b))
   - ☐ publication of the international application (under Rule 12.4(a))
   - ☐ international preliminary examination (under Rules 55.2(a) and/or 55.3(a) and (b))

2. With regard to the elements of the international application, this opinion has been established on the basis of *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this opinion as "originally filed")*:

   Description, Pages

   1-49 as originally filed

   Claims, Numbers

   1-42 filed with the demand for preliminary international examination

   Drawings, Sheets

   1/6-6/6 as originally filed

   ☐ a sequence listing - see Supplemental Box Relating to Sequence Listing.

3. ☐ The amendments have resulted in the cancellation of:
   - ☐ the description, pages
   - ☐ the claims, Nos.
   - ☐ the drawings, sheets/figs
   - ☐ the sequence listing *(specify)*:

4. ☒ This opinion has been established as if (some of) the amendments listed below had not been made, since either they are considered to go beyond the disclosure as filed, or they were not accompanied by a letter indicating the basis for the amendments in the application as filed, as indicated in the Supplemental Box (Rules 70.2(c) and (c-bis)):
   - ☐ the description, pages
   - ☒ the claims, Nos. 1-3, 38,39,41
   - ☐ the drawings, sheets/figs
   - ☐ the sequence listing *(specify)*:

5. ☐ This opinion has been established:
   - ☐ taking into account the rectification of an obvious mistake authorized by or notified to this Authority under Rule 91 (Rule 66.1(d-bis)).
   - ☐ without taking into account the rectification of an obvious mistake authorized by or notified to this Authority under Rule 91 (Rule 66.4bis).

6. ☐ Supplementary international search report(s) from Authority(ies) have been received and taken into account in drawing up this opinion (Rule 45bis.8(b) and (c)).

Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non- obvious), or to be industrially applicable have not been examined in respect of:

☐ the entire international application

☒ claims Nos. 4-37, 40, 42

because:

☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (specify):

☐ the description, claims or drawings *(indicate particular elements below)* or said claims Nos. are so unclear that no meaningful opinion could be formed *(specify)*:

☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed *(specify)*:

☒ no international search opinion has been established for the said claims Nos. 4-37, 40, 42

☐ a meaningful opinion could not be formed without sequence listing; the applicant did not, within the prescribed time limit:

☐ furnish a sequence listing on paper complying with the standard provided for in Annex C of the Administrative Instructions, and such listing was not available to the International Preliminary Examining Authority in a form and manner acceptable to it.

☐ furnish a sequence listing in electronic form complying with the standard provided for in Annex C of the Administrative Instructions, and such listing was not available to the International Preliminary Examining Authority in a form and manner acceptable to it.

☐ pay the required late furnishing fee for the furnishing of a sequence listing in response to an invitation under Rules 13*ter*.1(a) or (b) and 13*ter*.2.

☒ See supplemental sheet for further details

Box No. V Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	No:	Claims 1-5, 44, 45, 48
Inventive step (IS)	No:	Claims 1-5, 44, 45, 48
Industrial applicability (IA)	No:	Claims

2. Citations and explanations:

see separate sheet

Box No. VII Certain defects in the international application

The following defects in the form or contents of the international application have been noted:

see separate sheet

Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

Re Item I

Basis of the report

1 The International Preliminary Examining Authority considers that the following amendments to the claims go beyond the disclosure in the international application as filed:

In present claims 1 and 38, the "substrate material" is defined as "comprising at least one of diatomaceous earth, a clay, a zeolite, and a mineral". In paragraph [0061] of the description of the application as filed, however, only porous minerals are disclosed. No basis in the application as filed can be found for *any* mineral as "substrate material".

Further, in present claims 1 and 38, the "heat treatment" is defined as "comprising about 500°C to about 1200°C". In paragraph [0065] of the description of the application as filed, however, only each of the temperatures 500°C and 1200°C, respectively, are disclosed in an exemplified fashion. No basis in the application as filed can be found for a *temperature range from 500°C to 1200°C* of said "heat treatment".

Further, in present claim 1, the "promotion comprising chemical reaction with at least one of a halogen and a halide promotor" is defined as taking place "after the heat treatment". However, in none of the paragraphs [0058], [0061], [0065] or [0066] - [0074] mentioned in the applicant's letter as basis for this amendment said "promotion comprising chemical reaction with at least one of a halogen and a halide promotor" is specifically disclosed as taking place *after the heat treatment*.

The present opinion therefore has been established as if such amendments had not been made (Rule 70.2(c) PCT).

2 Claim 41 has been newly introduced. Since in the accompanying letter, the basis in the application as filed for this amendment was not indicated, this opinion has been established as if this amendment had not been made (Rule 70.2(c-bis).

3 Consequently, the present opinion has been established on the application documents as originally filed.

Re Item III

Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

4 No opinion was established with regard to novelty, inventive step and industrial applicability on the subject-matter of claims 4-37, 40 and 42 since their subject-matter relates to inventions for which no international search report has been established (Rule 66.2(e) PCT).

Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

Reference is made to the following documents:

D1 US 2011/168018 A1 (MOHAMADALIZADEH ALI [IR] ET AL) 14 July 2011 (2011-07-14)

D2 US 2009/136401 A1 (YANG XIAOLIN DAVID [US] ET AL) 28 May 2009 (2009-05-28)

D3 US 2004/076570 A1 (JIA CHARLES Q [CA]) 22 April 2004 (2004-04-22)

D4 A. Bakandritsos ET AL: "High Surface Area Montmorillonite-Carbon Composites and Derived Carbons", Chemistry of materials, vol. 16, no. 8, 16 March 2004 (2004-03-16), pages 1551-1559, XP055072551, ISSN: 0897-4756, DOI: 10.1021/cm0350030

D5 US 2008/090951 A1 (MAO DONGSHENG [US] ET AL) 17 April 2008 (2008-04-17)

5 For the reasons mentioned under Item I (see above), the present reasoned statement with regard to novelty, inventive step or industrial applicability has been established on the first (group of) inventions mentioned in the claims as originally filed. The claim numbering relates to those said claims as originally filed:

6 The present application does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 1-5, 44, 45 and 48 is not new in the sense of Article 33(2) PCT:

7 **D1** discloses a method for separating hydrogen sulfide (a pollutant) from a gas comprising methane, helium and hydrogen sulfide, the method comprising

providing a carbon nanocomposite sorbent of multiwall carbon nanotubes (MWCNT) with average diameters of from about 2 nm to about 5 nm and mesoporous crystalline material (MCM)-41;

contacting the said carbon nanocomposite sorbent with a flow of the gas containing 5500 ppm of hydrogen sulfide;

passing the gas flow to an outlet to give a separated gas containing 600 ppm of hydrogen sulfide (paragraph [0029]; paragraph [0062]; example 2).

D1 further discloses a method of making a carbon nanocomposite sorbent and the carbon nanocomposite sorbent hence made, the method comprising

providing MWCNT as carbon precursor;

providing MCM-41 as substrate material;

mixing MWCNTs and MCM-41 and thereby contacting them;

heating the mixture for 3 hours at 110°C (paragraph [0029]; paragraph [0062]; example 2).

D1 hence discloses the subject-matter of claims 1, 2, 44, 45 and 48.

8 **D2** discloses discloses a method for separating mercury from a flue gas comprising flue gas, fly ash and mercury, the method comprising

providing as-mined kaolin containing about 2% naturally intercalated carbon (Kaolin-1), which is considered as a carbon nanocomposite sorbent;

promoting the said carbon nanocomposite sorbent by impregnating it with a sodium bromide (NaBr) solution in water;

contacting the NaBr-promoted carbon nanocomposite sorbent with a flow of the gas containing mercury for one second by injecting the carbon nanocomposite sorbent into the gas in a drop tube reactor;

separating the NaBr-promoted carbon nanocomposite sorbent from the gas for analysis of mercury content (paragraph [0044] - paragraph [0047]; examples 8, 16; tables 1, 2; paragraph [0048] - paragraph [0050]; table 3).

D2 further discloses as-mined kaolin containing about 2% naturally intercalated carbon (Kaolin-1), which is considered as a carbon nanocomposite sorbent (paragraph [0044] - paragraph [0047]; examples 8; table 1).

D2 hence discloses the subject-matter of claims 1-5, 45 and 48.

9 **D3** discloses a method of separating mercury from a desiccator atmosphere that comprises mercury, the method comprising

providing treated coke (ECOcarbon), which is considered a carbon nanocomposite comprising agglomerated carbon structures of about 30 nm;

contacting the said carbon nanocomposite sorbent with a desiccator atmosphere that comprises mercury;

separating the said carbon nanocomposite sorbent comprising mercury from the desiccator atmosphere for analysis of mercury content (paragraph [0084] - paragraph [0086]; example 1; paragraph [0095] - paragraph [0099]; example 5).

D3 further discloses a method of making a carbon nanocomposite sorbent and the carbon nanocomposite sorbent hence made, the method comprising

providing fluid coke as carbon precursor;

providing sulfur dioxide (SO_2) as substrate material;

contacting fluid coke and SO_2 for 10 seconds;

heating the mixture at 800°C (paragraph [0087] - paragraph [0092]; example 2).

D3 hence discloses the subject-matter of claims 1-3, 44, 45 and 48.

10 **D4** discloses a method of making a carbon nanocomposite sorbent and the carbon nanocomposite sorbent hence made, the method comprising

providing sucrose as carbon precursor;

providing montmorillonite as substrate material;

contacting contacting sucrose and montmorillonite;

pyrolyzing and hence heating the mixture for one hour at 600 °C (Section: "Sample Preparation";page 1552, right-hand column; figure 1).

D4 hence discloses the subject-matter of claims 44, 45 and 48.

11 D5 discloses a method of making a carbon nanocomposite suitable as a sorbent and the carbon nanocomposite hence made, the method comprising

providing carbon nanotubes (CNTs) as carbon precursors,

providing epoxy as substrate material,

curing and hence heating the material for two hours at 160 °C .

D5 hence discloses the subject-matter of claims 44, 45 and 48 (paragraph [0015]; figure 1).

Re Item VII

Certain defects in the international application

12 Contrary to the requirements of Rule 5.1(a)(ii) PCT, the relevant background art documents mentioned in paragraphs [0005] to [0007] of the description are not cited in a way that allows their retrieval.

Re Item VIII

Certain observations on the international application

13 For the reasons mentioned under item I (see above), the present observations have been made with regard to the first (group of) inventions mentioned in the claims as originally filed. The claim numbering relates to those said claims as originally filed:

14 The application does not meet the requirements of Article 6 PCT for the following reasons:

15 The term "carbon nanocomposite" use in the claims is unclear, because it cannot be determined without ambiguity which further technical feature is meant to be attributed to the carbon composite by defining it as a "nano"composite. The term "nano" merely indicates a prefix in the SI and other systems of units denoting a factor of 10^{-9}. For the present purposes, any carbon composite is considered to be a "carbon nanocomposite". Insofar as

the description of the application contains a clear and unambiguous definition of the tem "carbon nanocomposite", said definition would have to be included in the claims to render the subject-matter of said claim clear.

16 The term pollutant used in claim 2 is vague and unclear and leaves the reader in doubt as to the meaning of the technical feature to which it refers. Specifically, whether a compound or material is considered to be polluting or not, depends on the specific circumstances in which the compound or material is found. Since theses specific circumstances are not further defined in the claim, the said claim is unclear. Insofar as the description of the application contains a clear and unambiguous definition of the tem "pollutant", said definition would have to be included in the claims to render the subject-matter of said claim clear.

17 Claim 4 is not supported by the description as required by Article 6 PCT, as its scope is broader than justified by the description and drawings. Specific support for "promoters" can only be found for "halogen and halide" promoters (see e.g. paragraph [0009] of the description), not for any "promoter".

Claim 48 tries to define the claimed carbon nanocomposite sorbent further in that it is "in contact with mercury or oxidized mercury". This wording introduces an ambiguity as to the scope of said claim 48. The expression "in contact" seeks to define a relationship between the entities "carbon nanocomposite sorbent" and "mercury or oxidized mercury", while it cannot be determined whether one of the two or both entities fall under the scope of the claim. The subject-matter of claim 48 therefore is unclear. For the present purposes of search and examination, claim 48 is considered to be solely directed to the "carbon nanocomposite sorbent" of claim 45, upon which it depends.

PATENT COOPERATION TREATY

From the
INTERNATIONAL SEARCHING AUTHORITY

To:

see form PCT/ISA/220

PCT

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

(PCT Rule 43*bis*.1)

Date of mailing
(day/month/year) see form PCT/ISA/210 (second sheet)

Applicant's or agent's file reference
see form PCT/ISA/220

FOR FURTHER ACTION
See paragraph 2 below

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/US2013/036964	17.04.2013	23.04.2012

International Patent Classification (IPC) or both national classification and IPC
INV. B01J20/12 B01D53/02 B01J20/30 B01J20/32 B01D53/64 B01J20/20

Applicant
ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION

1. This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☐ Box No. II Priority
- ☒ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☒ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Rule 43*bis*.1(a)(i) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☒ Box No. VII Certain defects in the international application
- ☒ Box No. VIII Certain observations on the international application

2. **FURTHER ACTION**

If a demand for international preliminary examination is made, this opinion will usually be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA") except that this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notifed the International Bureau under Rule 66.1*bis*(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of 3 months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

Name and mailing address of the ISA:	Date of completion of this opinion	Authorized Officer
European Patent Office P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk - Pays Bas Tel. +31 70 340 - 2040 Fax: +31 70 340 - 3016	see form PCT/ISA/210	Klemps, Christian Telephone No. +31 70 340-8036

Form PCT/ISA/237 (Cover Sheet) (July 2009)

Box No. I Basis of the opinion

1. With regard to the **language**, this opinion has been established on the basis of:

   ☒ the international application in the language in which it was filed

   ☐ a translation of the international application into , which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1 (b)).

2. ☐ This opinion has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43bis.1(a))

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, this opinion has been established on the basis of a sequence listing filed or furnished:

   a. (means)

      ☐ on paper

      ☐ in electronic form

   b. (time)

      ☐ in the international application as filed

      ☐ together with the international application in electronic form

      ☐ subsequently to this Authority for the purposes of search

4. ☐ In addition, in the case that more than one version or copy of a sequence listing has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.

5. Additional comments:

Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non obvious), or to be industrially applicable have not been examined in respect of

☐ the entire international application

☒ claims Nos. 6-43, 46, 47, 49

because:

☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international search *(specify)*:

☐ the description, claims or drawings *(indicate particular elements below)* or said claims Nos. are so unclear that no meaningful opinion could be formed *(specify)*:

☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed *(specify)*:

☒ no international search report has been established for the whole application or for said claims Nos. 6-43, 46, 47, 49

☐ a meaningful opinion could not be formed without the sequence listing; the applicant did not, within the prescribed time limit:

- ☐ furnish a sequence listing on paper complying with the standard provided for in Annex C of the Administrative Instructions, and such listing was not available to the International Searching Authority in a form and manner acceptable to it.
- ☐ furnish a sequence listing in electronic form complying with the standard provided for in Annex C of the Administrative Instructions, and such listing was not available to the International Searching Authority in a form and manner acceptable to it.
- ☐ pay the required late furnishing fee for the furnishing of a sequence listing in response to an invitation under Rules 13*ter*.1(a) or (b).

☐ See Supplemental Box for further details

Box No. IV Lack of unity of invention

1. ☒ In response to the invitation (Form PCT/ISA/206) to pay additional fees, the applicant has, within the applicable time limit:

   ☐ paid additional fees

   ☐ paid additional fees under protest and, where applicable, the protest fee

   ☐ paid additional fees under protest but the applicable protest fee was not paid

   ☒ not paid additional fees

2. ☐ This Authority found that the requirement of unity of invention is not complied with and chose not to invite the applicant to pay additional fees.

3. This Authority considers that the requirement of unity of invention in accordance with Rule 13.1, 13.2 and 13.3 is

   ☐ complied with

   ☒ not complied with for the following reasons:

   see separate sheet

4. Consequently, this report has been established in respect of the following parts of the international application:

   ☐ all parts.

   ☒ the parts relating to claims Nos. 1-5, 44, 45, 48

Box No. V Reasoned statement under Rule 43*bis*.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	
	No:	Claims	1-5, 44, 45, 48
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1-5, 44, 45, 48
Industrial applicability (IA)	Yes:	Claims	1-5, 44, 45, 48
	No:	Claims	

2. Citations and explanations

see separate sheet

Box No. VII Certain defects in the international application

The following defects in the form or contents of the international application have been noted:

see separate sheet

Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

Reference is made to the following documents:

D1 US 2011/168018 A1 (MOHAMADALIZADEH ALI [IR] ET AL) 14 July 2011 (2011-07-14)

D2 US 2009/136401 A1 (YANG XIAOLIN DAVID [US] ET AL) 28 May 2009 (2009-05-28)

D3 US 2004/076570 A1 (JIA CHARLES Q [CA]) 22 April 2004 (2004-04-22)

D4 A. Bakandritsos ET AL: "High Surface Area Montmorillonite-Carbon Composites and Derived Carbons", Chemistry of materials, vol. 16, no. 8, 16 March 2004 (2004-03-16), pages 1551-1559, XP055072551, ISSN: 0897-4756, DOI: 10.1021/cm0350030

D5 US 2008/090951 A1 (MAO DONGSHENG [US] ET AL) 17 April 2008 (2008-04-17)

Re Item IV

Lack of unity of invention

1 The present application lacks unity of invention (Rule 13 PCT) for the following reasons:

The present application contains the following eight independent claims: claim 1 (method claim), claim 6 (method claim), claim 43 (method claim), claim 44 (method claim), claim 45 (product claim), claim 46 (product claim), claim 47 (product claim), claim 49 (product claim).

The common same or corresponding technical features among those claims are:

carbon nanocomposite sorbent; method where said carbon nanocomposite sorbent is employed with a gas; said method comprising: the step of providing or obtaining a carbon nanocomposite sorbent; the step of contacting at least part of the sorbent; the step of separating from gas.

The common same or corresponding technical feature "carbon nanocomposite" is unclear (Article 6 PCT), because it cannot be determined which further technical feature is meant to be attributed to the carbon composite by defining it as a "nano"composite. The term "nano" merely

indicates a prefix in the SI and other systems of units denoting a factor of 10^{-9}. For the present purposes, any carbon composite is considered to be a "carbon nanocomposite".

The document US 2011/0168018 A1 (**D1**) discloses a method for separating hydrogen sulfide from a gas comprising methane, helium and hydrogen sulfide, the method comprising

providing a carbon nanocomposite sorbent of multiwall carbon nanotubes (MWCNT) with average diameters of from about 2 nm to about 5 nm and mesoporous crystalline material (MCM)-41;

contacting the said carbon nanocomposite sorbent with a flow of the gas containing 5500 ppm of hydrogen sulfide;

passing the gas flow to an outlet to give a separated gas containing 600 ppm of hydrogen sulfide (paragraph [0029]; paragraph [0062]; example 2).

Further, the document US 2009/0136401 A1 (**D2**) discloses a method for separating mercury from a flue gas comprising flue gas, fly ash and mercury, the method comprising

providing as-mined kaolin containing about 2% naturally intercalated carbon (Kaolin-1), which is considered as a carbon nanocomposite sorbent;

promoting the said carbon nanocomposite sorbent by impregnating it with a sodium bromide (NaBr) solution in water;

contacting the NaBr-promoted carbon nanocomposite sorbent with a flow of the gas containing mercury for one second by injecting the carbon nanocomposite sorbent into the gas in a drop tube reactor;

separating the NaBr-promoted carbon nanocomposite sorbent from the gas for analysis of mercury content (paragraph [0044] - paragraph [0047]; examples 8, 16; tables 1, 2; paragraph [0048] - paragraph [0050]; table 3).

All common same or corresponding technical features are known from **D1** and **D2**. These features therefore cannot serve as "special" technical features within the meaning of Rule 13.2 PCT.

Therefore, there is no common, same or corresponding special technical feature among all independent claims and thus the requirement of Rule 13.2 PCT is not met.

Amongst the independent claims 6, 43, 46, 47 and 49, the further common same or corresponding technical features are:

promoter; providing a halogen or halide promoter; promoting at least a portion of the carbon nanocomposite sorbent material by chemically reacting a (the) portion of the sorbent material with the halogen or halide promoter; promoted sorbent.

All common same or corresponding technical features are known from **D2**. These features therefore cannot serve as "special" technical features within the meaning of Rule 13.2 PCT.

Therefore, there is no common, same or corresponding special technical feature among all independent claims 6, 43, 46, 47 and 49 and thus the requirement of Rule 13.2 PCT is not met.

The separate groups of inventions are:

Group 1: claims: 1-5, 44, 45, 48 related to a method for separating a material from a gas that comprises the material, the method comprising providing or obtaining a carbon nanocomposite sorbent, contacting at least some of the material with the sorbent, to form a material-sorbent composition, and separating at least some of the material-sorbent composition from the material-containing gas, to give a separated gas; further to a method of making a such carbon nanocomposite sorbent; further to a such carbon nanocomposite.

Group 2: claims: 6-42, 46, 49 related to a method for reducing the pollutant content of a pollutant-containing gas, the method comprising providing or obtaining a carbon nanocomposite sorbent material, providing or obtaining a halogen or halide promotor, promoting at least a portion of the sorbent material by chemically reacting the portion of the sorbent material with the halogen or halide promoter to form a promoted sorbent, contacting at least part of the promoted sorbent with the pollutant in the pollutant-containing gas, to form a pollutant-sorbent composition, and separating particulates from the pollutant-containing gas, the particulates comprising at least some of the pollutant-sorbent composition, to give a cleaned gas; further to a promoted corbon nanocomposite sorbent made by reacting a portion of the sorbent material with a halogen or halide promoter to form the promoted sorbent.

Group 3: claims: 43, 47 related to a method for reducing the mercury content of a mercury-containing gas, the method comprising providing or obtaining a carbon nanocomposite sorbent, the carbon nanocomposite sorbent made by steps comprising providing or obtaining a carbon precursor,

providing or obtaining a substrate material, contacting the carbon precursor and the substrate material, to provide a nanocomposite starting material, and heating the nanocomposite starting material, to provide the carbon nanocomposite sorbent, providing or obtaining a halogen or halide promoter, promoting at least a portion of the sorbent material by chemically reacting the portion of the sorbent material with the halogen or halide promoter to form a promoted sorbent, contacting at least part of the promoted sorbent with the mercury, to form a mercury-sorbent composition, and separating at least some of the mercury-sorbent composition from the mercury-containing gas; further to a mercury-sorbent composition made by contacting a carbon nanocomposite sorbent with a mercury-containing gas.

The common problem underlying groups 1, 2 and 3 of inventions is seen to lie in the provision of a method for the removal of a (pollutant) component of a gas from said gas and the provision of a sorbent for a such removal.

The common problem is already known from both **D1** (see e.g. abstract) and **D2** (see e.g. abstract). No additional problem could be identified common to these three groups of inventions which could serve as the general inventive concept as required by Rule 13.1 PCT.

The application hence does not meet the requirements of unity of invention as defined in Rule 13.1 and 13.2 PCT.

In accordance with Article 17(3) PCT the international search report is established on those parts of the international application which relate to the invention first mentioned in the claims, i.e. the above mentioned Group 1.

Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

2 The present application further does not meet the criteria of Article 33(1) PCT, because the subject-matter of claims 1-5, 44, 45 and 48 is not new in the sense of Article 33(2) PCT:

3 **D1** discloses a method for separating hydrogen sulfide (a pollutant) from a gas comprising methane, helium and hydrogen sulfide, the method comprising

providing a carbon nanocomposite sorbent of multiwall carbon nanotubes (MWCNT) with average diameters of from about 2 nm to about 5 nm and mesoporous crystalline material (MCM)-41;

contacting the said carbon nanocomposite sorbent with a flow of the gas containing 5500 ppm of hydrogen sulfide;

passing the gas flow to an outlet to give a separated gas containing 600 ppm of hydrogen sulfide (paragraph [0029]; paragraph [0062]; example 2).

D1 further discloses a method of making a carbon nanocomposite sorbent and the carbon nanocomposite sorbent hence made, the method comprising

providing MWCNT as carbon precursor;

providing MCM-41 as substrate material;

mixing MWCNTs and MCM-41 and thereby contacting them;

heating the mixture for 3 hours at 110°C (paragraph [0029]; paragraph [0062]; example 2).

D1 hence discloses the subject-matter of claims 1, 2, 44, 45 and 48.

4 **D2** discloses discloses a method for separating mercury from a flue gas comprising flue gas, fly ash and mercury, the method comprising

providing as-mined kaolin containing about 2% naturally intercalated carbon (Kaolin-1), which is considered as a carbon nanocomposite sorbent;

promoting the said carbon nanocomposite sorbent by impregnating it with a sodium bromide (NaBr) solution in water;

contacting the NaBr-promoted carbon nanocomposite sorbent with a flow of the gas containing mercury for one second by injecting the carbon nanocomposite sorbent into the gas in a drop tube reactor;

separating the NaBr-promoted carbon nanocomposite sorbent from the gas for analysis of mercury content (paragraph [0044] - paragraph [0047]; examples 8, 16; tables 1, 2; paragraph [0048] - paragraph [0050]; table 3).

D2 further discloses as-mined kaolin containing about 2% naturally intercalated carbon (Kaolin-1), which is considered as a carbon nanocomposite sorbent (paragraph [0044] - paragraph [0047]; examples 8; table 1).

D2 hence discloses the subject-matter of claims 1-5, 45 and 48.

5 **D3** discloses a method of separating mercury from a desiccator atmosphere that comprises mercury, the method comprising

providing treated coke (ECOcarbon), which is considered a carbon nanocomposite comprising agglomerated carbon structures of about 30 nm;

contacting the said carbon nanocomposite sorbent with a desiccator atmosphere that comprises mercury;

separating the said carbon nanocomposite sorbent comprising mercury from the desiccator atmosphere for analysis of mercury content (paragraph [0084] - paragraph [0086]; example 1; paragraph [0095] - paragraph [0099]; example 5).

D3 further discloses a method of making a carbon nanocomposite sorbent and the carbon nanocomposite sorbent hence made, the method comprising

providing fluid coke as carbon precursor;

providing sulfur dioxide (SO_2) as substrate material;

contacting fluid coke and SO_2 for 10 seconds;

heating the mixture at 800°C (paragraph [0087] - paragraph [0092]; example 2).

D3 hence discloses the subject-matter of claims 1-3, 44, 45 and 48.

6 **D4** discloses a method of making a carbon nanocomposite sorbent and the carbon nanocomposite sorbent hence made, the method comprising

providing sucrose as carbon precursor;

providing montmorillonite as substrate material;

contacting contacting sucrose and montmorillonite;

pyrolyzing and hence heating the mixture for one hour at 600°C (Section: "Sample Preparation"; page 1552, right-hand column; figure 1).

D4 hence discloses the subject-matter of claims 44, 45 and 48.

7 **D5** discloses a method of making a carbon nanocomposite suitable as a sorbent and the carbon nanocomposite hence made, the method comprising

providing carbon nanotubes (CNTs) as carbon precursors,

providing epoxy as substrate material,

curing and hence heating the material for two hours at 160°C .

D5 hence discloses the subject-matter of claims 44, 45 and 48 (paragraph [0015]; figure 1).

Re Item VII

Certain defects in the international application

8 Contrary to the requirements of Rule 5.1(a)(ii) PCT, the relevant background art documents mentioned in paragraphs [0005] to [0007] of the description are not cited in a way that allows their retrieval.

Re Item VIII

Certain observations on the international application

9 The application does not meet the requirements of Article 6 PCT for the following reasons:

10 The term "carbon nanocomposite" use in the claims is unclear, because it cannot be determined without ambiguity which further technical feature is meant to be attributed to the carbon composite by defining it as a "nano"composite. The term "nano" merely indicates a prefix in the SI and other systems of units denoting a factor of 10^9. For the present purposes, any carbon composite is considered to be a "carbon nanocomposite".

11 The term pollutant used in claim 2 is vague and unclear and leaves the reader in doubt as to the meaning of the technical feature to which it refers. Specifically, whether a compound or material is considered to be polluting or not, depends on the specific circumstances in which the compound or material is found. Since theses specific circumstances are not further defined in the claim, the said claim is unclear.

12 Claim 4 is not supported by the description as required by Article 6 PCT, as its scope is broader than justified by the description and drawings. Specific support for "promoters" can only be found for "halogen and halide" promoters (see e.g. paragraph [0009] of the description), not for any "promoter".

13 Claim 48 tries to define the claimed carbon nanocomposite sorbent further in that it is "in contact with mercury or oxidized mercury". This wording introduces an ambiguity as to the scope of said claim 48. The expression "in

contact" seeks to define a relationship between the entities "carbon nanocomposite sorbent" and "mercury or oxidized mercury", while it cannot be determined whether one of the two or both entities fall under the scope of the claim. The subject-matter of claim 48 therefore is unclear. For the present purposes of search and examination, claim 48 is considered to be solely directed to the "carbon nanocomposite sorbent" of claim 45, upon which it depends.

PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference	FOR FURTHER ACTION	see Form PCT/ISA/220 as well as, where applicable, item 5 below.
3338021WO1		
International application No.	International filing date (day/month/year)	(Earliest) Priority Date (day/month/year)
PCT/US2014/019316	3 March 2014 (03-03-2014)	6 March 2013 (06-03-2013)

Applicant

ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION

This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of 8 sheets.

☐ It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**

a. With regard to the language, the international search was carried out on the basis of:

☒ the international application in the language in which it was filed

☐ a translation of the international application into _____, which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b))

b. ☐ This international search report has been established taking into account the rectification of an obvious mistake authorized by or notified to this Authority under Rule 91 (Rule 43.6bis(a)).

c. ☐ With regard to any nucleotide and/or amino acid sequence disclosed in the international application, see Box No. I.

2. ☐ **Certain claims were found unsearchable** (See Box No. II)

3. ☒ **Unity of invention is lacking** (see Box No III)

4. With regard to the title,

☒ the text is approved as submitted by the applicant

☐ the text has been established by this Authority to read as follows:

5. With regard to the abstract,

☒ the text is approved as submitted by the applicant

☐ the text has been established, according to Rule 38.2, by this Authority as it appears in Box No. IV. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority

6. With regard to the drawings,

a. the figure of the drawings to be published with the abstract is Figure No. _____

☐ as suggested by the applicant

☐ as selected by this Authority, because the applicant failed to suggest a figure

☐ as selected by this Authority, because this figure better characterizes the invention

b. ☐ none of the figures is to be published with the abstract

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US2014/019916

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

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

see additional sheet

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying an additional fees, this Authority did not invite payment of additional fees.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

1-49(partially)

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.

☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.

☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2014/019916

A. CLASSIFICATION OF SUBJECT MATTER
INV. B01J20/20 B01D53/02 B01D53/10 B01J20/32 C01B31/08
ADD.

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)
B01J C01B B01D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
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.
X	YUANJING ZHENG ET AL: "Review of technologies for mercury removal from flue gas from cement production processes", PROGRESS IN ENERGY AND COMBUSTION SCIENCE, ELSEVIER SCIENCE PUBLISHERS, AMSTERDAM, NL, vol. 38, no. 5, 26 April 2012 (2012-04-26), pages 599-629, XP028415255, ISSN: 0360-1285, DOI: 10.1016/J.PECS.2012.05.001 [retrieved on 2012-05-25]	1-9, 16-25, 38,39, 48,49
Y	page 600, paragraph 1 page 615, paragraph 4.2.5.1 page 615, right-hand column, line 1 - line 4 "...by impregnation of nitrogen-containing polymers or pitches ..."; page 616, left-hand column 2nd paragraph;	5-9, 16-25, 38,39, 48,49

-/--

[X] Further documents are listed in the continuation of Box C. [X] See patent family annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
21 May 2014	13/10/2014

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, Fax: (+31-70) 340-3016	Pöhlmann, Robert

Form PCT/ISA/210 (second sheet) (April 2005)

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(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
	page 616, right-hand column page 622; figure 20 paragraph before 4.2.23; page 612, left-hand column	
X	DE 20 2012 003747 U1 (BLUECHER GMBH [DE]) 15 November 2012 (2012-11-15)	1-4
Y	claims 1, 25 paragraphs [0106], [0142], [0143]	5-9, 16-25, 38,39, 48,49

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/US2014/019916

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
DE 202012003747 U1	15-11-2012	DE 102012007392 A1	08-05-2013
		DE 202012003747 U1	15-11-2012
		EP 2776153 A1	17-09-2014
		WO 2013068060 A1	16-05-2013

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. claims: 1-49(partially)

Pollutant removal from a gas by contacting with an activated carbon sorbent comprising nitrogen and applying the method for gases with a sulfur(VI) concentration of more than 3 ppm by mole

---

2. claims: 50-67(completely); 1-49(partially)

Pollutant removal from a gas by contacting with an activated carbon sorbent comprising nitrogen and obtaining or providing the activated carbon sorbent comprises promoting an unpromoted carbon sorbent and/or obtaining the unpromoted carbon sorbent from a carbon precursor comprising nitrogen and/or the activated carbon sorbent comprising a stabilizing agent

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3. claims: 1-49(partially)

Pollutant removal from a gas by contacting with an activated carbon sorbent comprising nitrogen and adding an alkaline component into the pollutant containing gas

---

4. claims: 1-49(partially)

Pollutant removal from a gas by contacting with an activated carbon sorbent comprising nitrogen and regenerating the activated carbon sorbent

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PATENT COOPERATION TREATY

From the INTERNATIONAL SEARCHING AUTHORITY

PCT

To:
Madden, Robert B. Reg. No. 57,521
SCHWEGMAN, LUNDBERG & WOESSNER
P.O. Box 2938
Minneapolis, MN 55402-0938
ETATS-UNIS D'AMERIQUE

INVITATION TO PAY ADDITIONAL FEES AND, WHERE APPLICABLE, PROTEST FEE

(PCT Article 17(3)(a) and Rule 40.1 and 40.2(e))

Applicant's or agent's file reference 3338021WO1	Date of mailing *(day/month/year)* 30 May 2014 (30-05-2014)
International application No. PCT/US2014/019916	**PAYMENT DUE** within **ONE MONTH** from the above date of mailing
	International filing date *(day/month/year)* 3 March 2014 (03-03-2014)

Applicant

ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION

1. This International Searching Authority

(i) considers that there are 4 *(number of)* inventions claimed in the international application covered by the claims indicated on an extra sheet:

(ii) therefore considers that the international application does not comply with the requirements of unity of invention (Rules 13.1, 13.2 and 13.3) for the reasons indicated on an extra sheet:

(iii) [X] has carried out a partial international search (see Annex) [ ] will establish the international search report on those parts of the international application which relate to the invention first mentioned in claims Nos.:

see extra sheet

(iv) will establish the international search report on the other parts of the international application only if, and to the extent to which, additional fees are paid.

2. Consequently, the applicant is hereby invited to pay, within the time limit indicated above, the amount indicated below:

EUR 1.875,00 (Fee per additional invention) x 3 (number of additional inventions) = EUR 5.625,00 (currency/total amount of additional fees)

3. The applicant is informed that, according to Rule 40.2(a), the payment of any additional fee may be made under protest, i.e., a reasoned statement to the effect that the international application complies with the requirement of unity of invention or that the amount of the required additional fee is excessive, where applicable, subject to the payment of a protest fee.

Where the applicant pays additional fees under protest, the applicant is hereby invited, within the time limit indicated above, to pay a protest fee (Rule 40.2(e)) in the amount of EUR 865,00 *(currency/amount)*

Where the applicant has not, within the time limit indicated above, paid the required protest fee, the protest will be considered not to have been made and the International Searching Authority will so declare.

4. [ ] Claim(s) Nos. _____ have been found to be unsearchable under Article 17(2)(b) because of defects under Article 17(2)(a) and therefore have not been included with any invention.

Name and mailing address of the International Searching Authority	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk Tel. (+31-70) 340-2040 Fax: (+31-70) 340-3016	SLOAN, Mary Tel: +49 (0)89 2399-2608

Schwegman Lundberg & Woessner P.A.
JUN 09 2014

Form PCT/ISA/206 (April 2005)
Registered Mail

PAC ET AL. EXHIBIT 1023
RECEIVED
Page 569

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. claims: 1-49(partially)

Pollutant removal from a gas by contacting with an activated carbon sorbent comprising nitrogen and applying the method for gases with a sulfur(VI) concentration of more than 3 ppm by mole

---

2. claims: 50-67(completely); 1-49(partially)

Pollutant removal from a gas by contacting with an activated carbon sorbent comprising nitrogen and obtaining or providing the activated carbon sorbent comprises promoting an unpromoted carbon sorbent and/or obtaining the unpromoted carbon sorbent from a carbon precursor comprising nitrogen and/or the activated carbon sorbent comprising a stabilizing agent

---

3. claims: 1-49(partially)

Pollutant removal from a gas by contacting with an activated carbon sorbent comprising nitrogen and adding an alkaline component into the pollutant containing gas

---

4. claims: 1-49(partially)

Pollutant removal from a gas by contacting with an activated carbon sorbent comprising nitrogen and regenerating the activated carbon sorbent

---

The common concept linking together the groups is the subject-matter of claim 1 . This common concept is not new over D1.
D1 discloses a method of reducing the mercury content in a mercury-containing gas (see abstract), the method comprising obtaining or providing an activated carbon sorbent comprising activated carbon sorbent particles comprising nitrogen in a surface layer of the sorbent particles (see page 615, para. 4.2.5.1 and page 616, left column, "...by impregnation of nitrogen-containing polymers or pitches ..."), contacting a pollutant-containing gas with the activated carbon sorbent to form a pollutant-sorbent composition (implicitly disclosed as for a sorbent based removal process the pollutant has to be contacted with the sorbent, the pollutant thereby always forms a pollutant-sorbent composition), and separating at least some of the pollutant-sorbent composition from the pollutant-containing gas, to give a separated gas (see page 622, figure 20).
Hence, D1 discloses all the features of claim 1.

D1 further discloses

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- claim 3: the concentration of nitrogen in the surface layer of the sorbent particles to be higher than in a core of the particles (see page 615, right column, lines 1-4, nitrogenated function introduced on carbon surface, therefrom following a lower nitrogenous concentration in the core of the particles)
- claim 5, 6: the mercury-containing gas to comprise a sulfur(VI) concentration, like SO 3 (see page 616, right column, 2nd paragraph)

The differentiating features are
- Group 1: applying the method for gases with a sulfur(VI) concentration of more than 3 ppm by mole
- Group 2: obtaining or providing the activated carbon sorbent comprises promoting an unpromoted carbon sorbent and/or providing or obtaining the unpromoted carbon sorbent from a carbon precursor comprising nitrogen and/or the activated carbon sorbent comprising a stabilizing agent
- Group 3: adding an alkaline component into the pollutant containing gas
- Group 4: regenerating the activated carbon sorbent

with following resulting technical effects:
- Group 1: no surprising and unexpected technical effect distinguishable
- Group 2: further enhancement of the mercury sorption efficiency of the activated carbon sorbent comprising nitrogen
- Group 3: reduction of sulfuric compounds in the pollutant containing gas
- Group 4: saving financial means for sorbent

Thus the problems solved may be regarded as
- Group 1: finding an alternative mercury containing gas for which the method can be applied.
- Group 2: how to modify the method of D1 in order to further enhance the mercury sorption efficiency of the activated carbon sorbent comprising nitrogen
- Group 3: how to modify the method of D1 in order to reduce the sulfuric compounds in the pollutant containing gas
- Group 4: how to modify the method of D1 in order to save financial means for sorbent

The special technical features (R. 13.2 PCT) are
- Group 1: no special technical feature A as the alternative gas is considered to be obvious for the skilled person.
- Group 2: differentiating feature = special technical feature B as the feature is neither disclosed nor rendered obvious by D1 and therefore is considered to make a contribution over D1.
- Group 3: differentiating feature = special technical feature C as the feature is neither disclosed nor rendered obvious by D1 and therefore is considered to make a contribution over D1.
- Group 4: no special technical feature D as the regeneration and reuse of the sorbent is disclosed by D1 (see page 612, left column, paragraph before 4.2.2.3).

Groups of (alleged) Inventions
The groups of technical features do not solve any common technical problem. Thus the groups of claims are neither linked by the same nor by corresponding special technical features and define different inventions not linked by a single general inventive concept (Rule 13.1 and 13.2 PCT)

and therefore the requirement for unity of invention referred to in Rule 13 PCT is not fulfilled .

PAC ET AL. EXHIBIT 1023
Page 572

Annex to Form PCT/ISA/206
COMMUNICATION RELATING TO THE RESULTS
OF THE PARTIAL INTERNATIONAL SEARCH

International Application No
PCT/US2014/019916

1. The present communication is an Annex to the invitation to pay additional fees (Form PCT/ISA/206). It shows the results of the international search established on the parts of the international application which relate to the invention first mentioned in claims Nos.:
see 'Invitation to pay additional fees'.
2. This communication is not the international search report which will be established according to Article 18 and Rule 43.

3. If the applicant does not pay any additional search fees, the information appearing in this communication will be considered as the result of the international search and will be included as such in the international search report.

4. If the applicant pays additional fees, the international search report will contain both the information appearing in this communication and the results of the international search on other parts of the international application for which such fees will have been paid.

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	YUANJING ZHENG ET AL: "Review of technologies for mercury removal from flue gas from cement production processes", PROGRESS IN ENERGY AND COMBUSTION SCIENCE, ELSEVIER SCIENCE PUBLISHERS, AMSTERDAM, NL, vol. 38, no. 5, 26 April 2012 (2012-04-26) , pages 599-629, XP028415255, ISSN: 0360-1285, DOI: 10.1016/J.PECS.2012.05.001 [retrieved on 2012-05-25]	1-9, 16-25, 38,39, 48,49
Y	page 600, paragraph 1 page 615, paragraph 4.2.5.1 page 615, right-hand column, line 1 - line 4 "...by impregnation of nitrogen-containing polymers or pitches ..."; page 616, left-hand column 2nd paragraph; page 616, right-hand column page 622; figure 20 paragraph before 4.2.23; page 612, left-hand column	5-9, 16-25, 38,39, 48,49
X	DE 20 2012 003747 U1 (BLUECHER GMBH [DE]) 15 November 2012 (2012-11-15)	1-4
Y	claims 1, 25 paragraphs [0106], [0142], [0143]	5-9, 16-25, 38,39, 48,49

☐ 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

4

Form PCT/ISA/206 (Annex, first sheet) (July 1992; reprint January 2004)

Patent Family Annex

Information on patent family members

International Application No
PCT/US2014/019916

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
DE 202012003747 U1	15-11-2012	DE 102012007392 A1	08-05-2013
		DE 202012003747 U1	15-11-2012
		WO 2013068060 A1	16-05-2013

PATENT COOPERATION TREATY

From the
INTERNATIONAL SEARCHING AUTHORITY

To:

see form PCT/ISA/220

PCT

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY
(PCT Rule 43*bis*.1)

Date of mailing
(day/month/year) see form PCT/ISA/210 (second sheet)

Applicant's or agent's file reference
see form PCT/ISA/220

FOR FURTHER ACTION
See paragraph 2 below

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/US2014/019916	03.03.2014	06.03.2013

International Patent Classification (IPC) or both national classification and IPC
INV. B01J20/20 B01D53/02 B01D63/10 B01J20/32 C01B31/08

Applicant
ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION

1. This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☐ Box No. II Priority
- ☒ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☒ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Rule 43*bis*.1(a)(i) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☒ Box No. VII Certain defects in the international application
- ☒ Box No. VIII Certain observations on the international application

2. **FURTHER ACTION**

If a demand for international preliminary examination is made, this opinion will usually be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA") except that this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1*bis*(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of 3 months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

Name and mailing address of the ISA:	Date of completion of this opinion	Authorized Officer
European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Fax: +49 89 2399 - 4465	see form PCT/ISA/210	Pöhlmann, Robert Telephone No. +49 89 2399-2089

Form PCT/ISA/237 (Cover Sheet) (July 2009)

Box No. I Basis of the opinion

1. With regard to the **language**, this opinion has been established on the basis of:

   ☒ the international application in the language in which it was filed

   ☐ a translation of the international application into , which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1 (b)).

2. ☐ This opinion has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43bis.1(a))

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, this opinion has been established on the basis of a sequence listing filed or furnished:

   a. (means)

      ☐ on paper

      ☐ in electronic form

   b. (time)

      ☐ in the international application as filed

      ☐ together with the international application in electronic form

      ☐ subsequently to this Authority for the purposes of search

4. ☐ In addition, in the case that more than one version or copy of a sequence listing has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.

5. Additional comments:

Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non obvious), or to be industrially applicable have not been examined in respect of

☐ the entire international application

☒ claims Nos. 50-67(completely); 1-49(partially)

because:

☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international search *(specify)*:

☐ the description, claims or drawings *(indicate particular elements below)* or said claims Nos. are so unclear that no meaningful opinion could be formed *(specify)*:

☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed *(specify)*:

☒ no international search report has been established for the whole application or for said claims Nos. 50-67(completely); 1-49(partially)

☐ a meaningful opinion could not be formed without the sequence listing; the applicant did not, within the prescribed time limit:

- ☐ furnish a sequence listing on paper complying with the standard provided for in Annex C of the Administrative Instructions, and such listing was not available to the International Searching Authority in a form and manner acceptable to it.
- ☐ furnish a sequence listing in electronic form complying with the standard provided for in Annex C of the Administrative Instructions, and such listing was not available to the International Searching Authority in a form and manner acceptable to it.
- ☐ pay the required late furnishing fee for the furnishing of a sequence listing in response to an invitation under Rules 13*ter*.1(a) or (b).

☐ See Supplemental Box for further details

Box No. IV Lack of unity of invention

1. ☒ In response to the invitation (Form PCT/ISA/206) to pay additional fees, the applicant has, within the applicable time limit:

   ☐ paid additional fees

   ☐ paid additional fees under protest and, where applicable, the protest fee

   ☐ paid additional fees under protest but the applicable protest fee was not paid

   ☒ not paid additional fees

2. ☐ This Authority found that the requirement of unity of invention is not complied with and chose not to invite the applicant to pay additional fees.

3. This Authority considers that the requirement of unity of invention in accordance with Rule 13.1, 13.2 and 13.3 is

   ☐ complied with

   ☒ not complied with for the following reasons:

   see separate sheet

4. Consequently, this report has been established in respect of the following parts of the international application:

   ☐ all parts.

   ☒ the parts relating to claims Nos. 1-49(partially)

Box No. V Reasoned statement under Rule 43*bis*.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	5-49(partially)
	No:	Claims	1-4(partially)
Inventive step (IS)	Yes:	Claims	10-15, 26-37, 40-47(all partially)
	No:	Claims	1-9, 16-25, 38, 39, 48, 49(all partially)
Industrial applicability (IA)	Yes:	Claims	1-49(partially)
	No:	Claims	

2. Citations and explanations

   see separate sheet

Box No. VII Certain defects in the international application

The following defects in the form or contents of the international application have been noted:

see separate sheet

Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

Re Item VIII

Certain observations on the international application

1 Clarity, Article 6 PCT)

1.1 General

The terms "about" (used in claims 4-6, 18-24, 34, 50, 58, 64), "substantially" (used in claims 5, 7-9, 50, 61-64, 67) and "core" (used in claims 3, 7, 18, 19, 51, 56, 61) are vague and unclear and leave the reader in doubt as to the meaning of the technical features to which they refer, thereby rendering the definition of the subject-matter of said claims unclear, Art. 6 PCT.

1.2 Claim 4

Claim 4 does not meet the requirements of Article 6 PCT because the matter for which protection is sought is not clearly defined. The claim attempts to define the subject-matter in terms of the result to be achieved (combines with at least 70 wt%), which merely amounts to a statement of the underlying problem, without providing the technical features necessary for achieving this result (like but not limited to volume flows, distribution of sorbent, injection of sorbent, contact times, temperatures, other components in the mercury-containing gas).

1.3 Claim 7

It is unclear (Art. 6 PCT) how the nitrogen can be homogeneously distributed in the core of the particles and how at the same time there can be a concentration profile from the surface to the core (when referring to claim 3).

1.4 Claims 8, 9

Claims 8 and 9 do not meet the requirements of Art. 6 PCT because the matter for which protection is sought is not clearly defined. The claims attempt to define the subject-matter in terms of the result to be achieved, which merely amounts to a statement of the underlying problem, without providing the technical features necessary for achieving this result.

1.5 Claim 18

It is unclear in claim 18 how a core of an activated carbon sorbent particle can comprise 99 wt% nitrogen (Art. 6 PCT). The core practically would consist of nitrogen only in this case.

1.6 Claim 20

It is unclear in claim 20 how a surface layer of an activated carbon sorbent particle can comprise 99 wt% nitrogen (Art. 6 PCT). The surface would consist of nitrogen only in this cas. Also it is unclear if the technical effect related to nitrogen being present at 0.001 wt% in the surface layer is still existing (Art.6 PCT).

1.7 Claim 22

The definition of the surface layer is unclear as a thickness of 99% of the radius of the particles is in fact the complete particle and not only a "surface layer" (Art. 6 PCT).

Re Item IV

2 **Lack of unity of invention**

Notwithstanding the objections under Article 6 PCT above, the application lacks unity within the meaning of Rule 13.1 PCT.

2.1 Reference is made to the following document:

D1 YUANJING ZHENG ET AL: "Review of technologies for mercury removal from flue gas from cement production processes",PROGRESS IN ENERGY AND COMBUSTION SCIENCE, ELSEVIER SCIENCE PUBLISHERS, AMSTERDAM, NL,vol. 38, no. 5, 26 April 2012 (2012-04-26), pages 599-629, XP028415255,ISSN: 0360-1285, DOI: 10.1016/J.PECS.2012.05.001[retrieved on 2012-05-25]

D2 DE 20 2012 003747 U1 (BLUECHER GMBH [DE]) 15 November 2012 (2012-11-15)

2.2 **There are several groups of claims:**

- Group 1: Claims 1-49 (partially), in particular claims 2-9, 16-25, 38, 39, 48, 49
- Group 2: Claims 1-49 (partially), in particular claims 10-15, 26-37, 40, 41, 44, 45, claims 50-67 (completely)
- Group 3: Claims 1-49 (partially), in particular claims 42, 43
- Group 4: Claims 1-49 (partially), in particular claims claims 46, 47

2.3 **The common concept linking together the groups is the subject-matter of claim 1.** This common concept is not new over D1.

2.3.1 D1 discloses a method of reducing the mercury content in a mercury-containing gas (see page 600, para. 1.), the method comprising obtaining or providing an activated carbon sorbent comprising activated carbon sorbent particles comprising nitrogen in a surface layer of the sorbent particles (see page 615, para. 4.2.5.1 and page 616, left column, "...by impregnation of nitrogen-containing polymers or pitches ..."), contacting a pollutant-containing gas with the activated carbon sorbent to form a pollutant-sorbent composition (implicitly disclosed as for a sorbent based removal process the pollutant has to be contacted with the sorbent, the pollutant thereby always forms a pollutant-sorbent composition), and separating at least some of the pollutant-sorbent composition from the pollutant-containing gas, to give a separated gas (see page 622, figure 20).

Hence, D1 discloses all the features of claim 1.

2.3.2 D1 further discloses

- claim 3: the concentration of nitrogen in the surface layer of the sorbent particles to be higher than in a core of the particles (see page 615, right column, lines 1-4, nitrogenated function introduced on carbon surface, therefrom following a lower nitrogenous concentration in the core of the particles)

- claim 5, 6: the mercury-containing gas to comprise a sulfur(VI) concentration, like SO_3 (see page 616, right column, 2nd paragraph)

2.4 **The differentiating features are**

- Group 1: applying the method for gases with a sulfur(VI) concentration of more than 3 ppm by mole
- Group 2: obtaining or providing the activated carbon sorbent comprises promoting an unpromoted carbon sorbent and/or providing or obtaining the unpromoted carbon sorbent from a carbon precursor comprising nitrogen and/or the activated carbon sorbent comprising a stabilizing agent
- Group 3: adding an alkaline component into the pollutant containing gas
- Group 4: regenerating the activated carbon sorbent

2.5 **with following resulting technical effects:**

- Group 1: no surprising and unexpected technical effect distinguishable

- Group 2: further enhancement of the mercury sorption efficiency of the activated carbon sorbent comprising nitrogen
- Group 3: reduction of sulfuric compounds in the pollutant containing gas
- Group 4: saving financial means for sorbent

2.6 **Thus the problems solved may be regarded as**

- Group 1: finding an alternative mercury containing gas for which the method can be applied.
- Group 2: how to modify the method of D1 in order to further enhance the mercury sorption efficiency of the activated carbon sorbent comprising nitrogen
- Group 3: how to modify the method of D1 in order to reduce the sulfuric compounds in the pollutant containing gas
- Group 4: how to modify the method of D1 in order to save financial means for sorbent

2.7 **The special technical features (R. 13.2 PCT) are**

- Group 1: no special technical feature A as the alternative gas is considered to be obvious for the skilled person.
- Group 2: differentiating feature = special technical feature B as the feature is neither disclosed nor rendered obvious by D1 and therefore is considered to make a contribution over D1.
- Group 3: differentiating feature = special technical feature C as the feature is neither disclosed nor rendered obvious by D1 and therefore is considered to make a contribution over D1.
- Group 4: no special technical feature D as the regeneration and reuse of the sorbent is disclosed by D1 (see page 612, left column, paragraph before 4.2.2.3).

2.8 **Groups of (alleged) inventions**

The groups of technical features do not solve any common technical problem. Thus the groups of claims are neither linked by the same nor by corresponding special technical features and define different inventions not linked by a single general inventive concept (Rule 13.1 and 13.2 PCT) and therefore **the requirement for unity of invention referred to in Rule 13 PCT is not fulfilled**.

2.9 **Consequence**

A search has been performed for the invention first mentioned in the claims, here the subject-matter of the claims forming group 1. Correspondingly, in the following this written opinion is restricted to group 1.

Group 1 : Claims 1-9, 16-25, 38, 39, 48, 49 (partially)

Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

3 Novelty Claim 1

3.1 As already shown under point 2.3.1 above the subject-matter of claim 1 is not new (Article 33(2) PCT).

4 Inventive Step Claim 1

Besides D1, which is considered to be the closest prior art to the subject-matter of claim 1, also D2 could be regarded as the closest prior art the subject-matter of claim 1.

4.1.1 D2 discloses

A method of reducing the pollutant content in a pollutant-containing gas (see §0142 and 0143), the method comprising:

obtaining or providing an activated carbon sorbent comprising activated carbon sorbent particles comprising nitrogen in a surface layer of the sorbent particles (see claim 1);

4.1.2 The subject-matter of claim 1 therefore differs from this known D2 in that

a pollutant-containing gas is contacted with the activated carbon sorbent, to form a pollutant-sorbent composition; and in that at least some of the pollutant-sorbent composition is separated from the pollutant-containing gas, to give a separated gas.

4.1.3 There is no unexpected technical effect related to that difference.

4.1.4 The problem to be solved by the present invention may therefore be regarded as how to to apply the method of D2 for reducing the pollutant content in a pollutant-containing gas.

4.1.5 The solution proposed in claim 1 of the present application cannot be considered to involve an inventive step (Article 33(3) PCT) for the following reasons:

It is common general knowledge for the skilled person to contact the sorbent with the gas to form a pollutant-sorbent composition and to separate the pollutant-sorbent composition from the gas to give a separated gas. Also, amongst many other documents he finds in D1 an indication (see cited passages) with which he would contact the sorbent with the gas, thereby forming a pollutant sorbent composition and separate this composition from the gas to give a separated gas, thereby arriving at the subject-matter of claim 1 without being inventive.

5 Dependent claims

Dependent claims 2-9, 16-25, 38, 39, 48, 49 do not contain any features which, in combination with the features of any claim to which they refer, meet the requirements of the PCT in respect of novelty and/or inventive step.

5.1 Claims 2, 3

The subject-matter of said claims is neither new (see points 2.3.1 and 2.3.2 above), nor inventive (see D2, claim 1 and §0143, mercury being an obvious toxic substance whose removal from gas streams with activated carbon is widely known in industry).

5.2 Claims 5, 6

D1 discloses a sulfur(VI) concentration in the mercury-containing gas (see 2.3.2 above) and the claimed ranges for said concentration are considered to be merely an arbitrary selection out of a variety of possible mercury containing gases without showing an unexpected technical effect. The properties of the activated carbon sorbent of D1 are considered to achieve the results claimed in claim 5 with regard to corresponding activated carbon sorbents.

5.3 Claims 16, 17, 38, 39, 48, 49

It is well-known in the art to add the sorbent into the mercury-containing gas and to remove it after a certain distance by various means like bag houses, electrostatic precipitators, cyclones or the like. As well, it is common general knowledge to contact the gas with the sorbent in a fixed bed, a dry or aqueous scrubber, a filter or a moving bed.

Claim 19, 21

D2 discloses a wide variety of possible nitrogen concentrations of the activated carbon (see claim 25), which is also considered to be applicable either to the surface and/or to the core of carbon particles. Therefore, the claimed ranges are not inventive.

Claim 24, 25

The claimed range of particle diameters is considered an arbitrary selection and activated carbon particles in this range are known to the skilled person (see for reference D2, §0106). Furthermore, every surface is considered to be continuous and therefore also a surface layer.

Re Item VII

Certain defects in the international application

6 The requirements of Rule 6.3(b) PCT (two-part form of independent claims) and Rule 5.1(a)(ii) and (iii) PCT (acknowledgement of prior art D1) are not met.

Electronic Acknowledgement Receipt

EFS ID:	33295382
Application Number:	15951970
International Application Number:	
Confirmation Number:	6410
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Customer Number:	21186
Filer:	Jordan Rivard Golomb/Jamie Johnson
Filer Authorized By:	Jordan Rivard Golomb
Attorney Docket Number:	4820.001US8
Receipt Date:	26-JUL-2018
Filing Date:	12-APR-2018
Time Stamp:	16:21:28
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Non Patent Literature	0237_ep_response_05_11_10.pdf	596450 e848138ccaa4a7d7268f5e2175f0f8eda588f878	no	11

Warnings:

Information:					
2	Non Patent Literature	0238_4820001ep1_eesrr_090911.pdf	2247240 fe8845c219330394989db1f3fbb3e5472518 68e5	no	20
Warnings:					
Information:					
3	Non Patent Literature	0239_third_party_observations.pdf	674454 1aae9dcedc9f20d93f2cf0739e5acee12f862 f59	no	7
Warnings:					
Information:					
4	Non Patent Literature	0240_third_party_comment_12_04_07.pdf	1423810 de9a1f0e71899e26a598a15a979a39000f9f 6f59	no	5
Warnings:					
Information:					
5	Non Patent Literature	0241_4820007ep1_decisiontograntt_041714.pdf	239269 bf334ab5741e97b89738424f581eb1b97f4c 6409	no	2
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Information:					
6	Non Patent Literature	0242_4820007ep1_eesr_120612.pdf	509966 15d215dfaeef41b4be4c7a286d23e70f53ad 6aea	no	5
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Information:					
7	Non Patent Literature	0243_4820007ep1_e2000c_031714.pdf	7019813 068ccc85f5b2507abf1ab879d931c07e96f1 6a0f	no	44
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8	Non Patent Literature	0244_4820007ep1_e2000c_093013.pdf	6729751 07c5acbcc5ea5b50b8aaa64ec3bbe76f875 36e8a	no	44
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Information:					

9	Non Patent Literature	0245_4820007ep1_aarmisc_020314.pdf	1209447 96bc11afbd872340cf12cef52356c26e82028fab	no	11
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Information:					
10	Non Patent Literature	0246_4820007ep1_eesrr_062013.pdf	2165813 0e9a3f2825f7444be68532cfa7c5172178d001ec	no	18
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Information:					
11	Non Patent Literature	0247_3_4820001ep3_e2001_112612.pdf	325591 0b68f1483734e55391a5a142232698f8a96ad1f0	no	4
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12	Non Patent Literature	0248_7_4820001ep3_decision_121913.pdf	1394786 ac592e1528f70d798f4a73facb820ddd3f389940	no	18
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Information:					
14	Non Patent Literature	0250_6_4820001ep3_oar_misc_110413.pdf	260210 81fdad3e2f0916531809 24f1339e440004eeaaf9	no	4
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Information:					
15	Non Patent Literature	0251_4_4820001ep3_e2001r4_020413.pdf	926261 c88d6c17540c784e672f2b4090d45751242eb12d	no	9
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Information:					

16	Non Patent Literature	0252_2_4820001ep3_eesrr_082812.pdf	1062658 3b7968421d861c25940e7ab458b4c7d37f48eb48	no	9
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17	Non Patent Literature	0253_5_4820001ep3_oarmisc_041513.pdf	632129 1fbc3b4c86174694122afe74fc0d38c9e356033	no	7
Warnings:					
Information:					
18	Non Patent Literature	0254_4820001ep2_e2001_010913.pdf	351110 22bbf16f53ed114618a3f21fcf7f84d55dadca9e	no	4
Warnings:					
Information:					
19	Non Patent Literature	0255_4820001ep2_decisiontograntt_050214.pdf	240468 e26be6acaf71923ec49d315f30927175a383d360	no	2
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Information:					
20	Non Patent Literature	0256_4820001ep2_eesr_012312.pdf	571436 2f2de6b99e2ac0856616894e7a2ae2186cf2a501	no	6
Warnings:					
Information:					
21	Non Patent Literature	0257_4820001ep2_eesr_050912.pdf	1199398 cf80801c300523c3f7d5f777cc63b34f05603a8f	no	12
Warnings:					
Information:					
22	Non Patent Literature	0258_4820001ep2_e2001_103013.pdf	250916 65f4facf61e0c09e2ccc34eef68e5741bf9c13d4	no	4
Warnings:					
Information:					

23	Non Patent Literature	0259_4820001ep2_oarmisc_121113.pdf	609183 ced50c440bc978ca81236eb5fbacb1ead7339146	no	6
Warnings:					
Information:					
24	Non Patent Literature	0260_4820001ep2_e2001ar_021113.pdf	706120 834f5d386511f9d6ef9bd1cf84f48bfc87c64541	no	5
Warnings:					
Information:					
25	Non Patent Literature	0261_4820001ep2_aarmisc_101013.pdf	2039055 6c8bb12ab190fc30d213fe42c68a26a9906c05b5	no	18
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Information:					
26	Non Patent Literature	0262_4820001ep2_eesrr_112212.pdf	1553524 ef3445e718168e84c149aa066b56b13532ab7926	no	13
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Information:					
27	Non Patent Literature	0263_4820001ep2_oarmisc_041513.pdf	598635 c71f380fc0f4bb0f25b7cf4f02f0d672e2a369b0	no	6
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Information:					
28	Non Patent Literature	0264_4820006ep1_oarmisc_042417.pdf	655151 97b9eaae1b0e292d55b8489e0fcb871983647837	no	8
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29	Non Patent Literature	0265_3338016ep1_oarmisc_051217.pdf	702332 af8ee10637fc09bfb716db925db1dff9d34a7593	no	6
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30	Non Patent Literature	0266_ep_oa_12_19_2014.pdf	338900 98add43b5aaf29d7b66bf8d8755ac5d3e96dd1fe	no	2
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Information:					
31	Non Patent Literature	0267_ep_response_06_29_2015.pdf	5327099 45eae82be1a2817f7545614 65b990b43838acaee	no	19
Warnings:					
Information:					
32	Non Patent Literature	0268_3338021ep1_e2001_022217.pdf	655001 91701743c61b7325f093b79be6741ad056613914	no	6
Warnings:					
Information:					
33	Non Patent Literature	0269_147111066e2001r4_081717_20170222.pdf	146560 a6e55ebfd90fe40ebec43fe9b07e5c70bd51a065	no	10
Warnings:					
Information:					
34	Non Patent Literature	0270_2008071986_pct_13009.pdf	330236 21e049c586991a0201e14ac5abafb24794751f56	no	3
Warnings:					
Information:					
35	Non Patent Literature	0271_4820001wo1_p326_122706.pdf	462866 0724c502b8f84fd1b74a3a948b59381a028c19a4	no	3
Warnings:					
Information:					
36	Non Patent Literature	0272_4820001wo1_p210_072506.pdf	174185 8c71b66231e45d204fe2f08d1257a9560e4b3344	no	2
Warnings:					
Information:					

37	Non Patent Literature	0273_4820001wo1_p237_072506.pdf	351881 1d10c5e4a9a4f1335e57fe9ed380b91fb47d9c19	no	3
Warnings:					
Information:					
38	Non Patent Literature	0274_4820007wo1_p326_110311.pdf	623895 2efc4a6be3d7ec8a2496ef328c9603b03fca4f9b	no	6
Warnings:					
Information:					
39	Non Patent Literature	0275_4820007wo1_p210_091310.pdf	350307 258f00ec157f395f321115dfdfcf9c11be741356	no	3
Warnings:					
Information:					
40	Non Patent Literature	0276_4820007wo1_p237_091310.pdf	453772 06fb741fe0cc89c5097f25c701c8c671cf1bc8b1	no	4
Warnings:					
Information:					
41	Non Patent Literature	0277_3338016wo1_p326_071714.pdf	1867110 f1a8de3b77b3141cadcb532b8b60e90dcf308e1b	no	11
Warnings:					
Information:					
42	Non Patent Literature	0278_3338016wo1_p210_112913.pdf	804642 040fc536e7cd18769a691ada2c6734af97ff7466	no	7
Warnings:					
Information:					
43	Non Patent Literature	0279_3338016wo1_p206_080213.pdf	1096404 0446e53f77c93c574ef4f41efed9e54904564160	no	7
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Information:					

44	Non Patent Literature	0280_3338016wo1_p237_041514.pdf	1414324 c0c95ea8c3b4e119d690d5024a08a6d738c04f31	no	10
Warnings:					
Information:					
45	Non Patent Literature	0281_3338016wo1_p237_112913.pdf	1600046 2b647c51701e275d31589800ae33eeeae25a93f2	no	13
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46	Non Patent Literature	0282_inter_sr_10_13_2014.pdf	1479526 3a58ab5369f842acee9e5de8920fa6ca8a1d8ae1	no	6
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Information:					
47	Non Patent Literature	0283_3338021wo1_p206_053014.pdf	967790 7aadd94d1b7d519bcfd0d861225f76b69d4c8d7e	no	6
Warnings:					
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48	Non Patent Literature	0284_written_opinion_10_13_2014.pdf	3453405 9a4a2bcef5f83f4f27aba8a85626c3ffdffb554c	no	12
Warnings:					
Information:					
49	Non Patent Literature	0285_powdered_activated_carbon___cabot_corporation.pdf	469745 70d429c8a93f692930b99b21ae758bb27f01e4dc	no	3
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Information:					
50	Non Patent Literature	0286_137193389aarmisc_090517_201709042.pdf	109442 5a1b2e21236b656041589f8a4a215317c3f2a654	no	7
Warnings:					
Information:					

51	Non Patent Literature	0287_supp_eu_search_rpt_126 2012.pdf	222511 6e0e474afe9910be599b154992a0b96c49e 1a059	no	2
Warnings:					
Information:					
52	Non Patent Literature	0288_14195360_co2_removal_ potential_of_carbons_prepare d.pdf	1515182 7711e4564863441eb43236fc87685986236 59bd2	no	9
Warnings:					
Information:					
53	Non Patent Literature	0289_bakandritos_2005.pdf	290716 fac131ecd8e9549c898384bd0dd9bbd8d95 a5d9c	no	7
Warnings:					
Information:					
54	Non Patent Literature	0290_bakandritsos_2004.pdf	814947 e0b014df8de463094125360490016288803e 4c619	no	9
Warnings:					
Information:					
55	Non Patent Literature	0291_141953602.pdf	1620503 ba520585f59a8fac00563aaf0663c835b89e 16c2	no	7
Warnings:					
Information:					
56	Non Patent Literature	0292_chang_2004.pdf	5359338 33bd2c8a2d2413a16b9639f56c8b99d3ada 80684	no	9
Warnings:					
Information:					
57	Non Patent Literature	0293_criswell.pdf	4825088 7bb1a762d7097b98e2eb30758101a355ee 637651	no	23
Warnings:					
Information:					

58	Non Patent Literature	0294_darder_ruiz_hitzky_2005.pdf	6086924 4a8f9e3ef486e76b9514d96fb4ac3fbf4f7b9fb2	no	6
Warnings:					
Information:					
59	Non Patent Literature	0295_14195360_sorption_of_mercury_by_activated_carbon.pdf	1284203 3a44011182446d2f50b584fc2b31d74f6a3da4bf	no	6
Warnings:					
Information:					
60	Non Patent Literature	0296_dong_2009.pdf	7505144 3c04b677c8b55c5f493a8daa9f76ebb3e457a247	no	6
Warnings:					
Information:					
		Total Files Size (in bytes):	89638025		

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.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US04/12828

A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) : B01J 20/34, 38/48, 38/60, 38/62
US CL : 502/22, 27, 28

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)
U.S. : 502/22, 27, 28

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
NONE

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
Please See Continuation Sheet

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X, P --- Y,P	US 6,579,507 B2 (PAHLMAN et al) 17 June 2003 (17.06.2003), column 10, lines 39-67; column 40, lines 32-67.	1-3,11-14,17-22,26-32,36-38,42-48,55-58, and 62-69 --- 4-10,15,16,23-25,33-35,39-41,49-51, and 59-61
X	US 6,475,451 B1 (LEPPIN et al) 05 November 2002 (05.11.2002), see abstract.	43-48
A	US 5,827,352 (ALTMAN et al) 27 October 1998 (27.10.1998), see abstract and entire patent.	1-69
Y	US 5,480,619 (JOHNSON et al) 02 January 1996 (02.01.1996), see abstract, col . 4, lines 30-35.	7-10, 15,16,23-25,33-35,39-41,49-51 and 59-61
Y	US 4,196,173 (deJong et al.) 01 April 1980 (01.04.1980), see abstract.	4-6

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent 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
22 September 2004 (22.09.2004)

Date of mailing of the international search report
22 OCT 2004

Name and mailing address of the ISA/US
Mail Stop PCT, Attn: ISA/US
Commissioner of Patents
P.O. Box 1450
Alexandria, Virginia 22313-1450
Facsimile No. (703) 305-3230

Authorized officer
Jonas N. Strickland
Telephone No. 571-272-1700

Form PCT/ISA/210 (second sheet) (July 1998)

Continuation of B. FIELDS SEARCHED Item 3:
EAST, WEST search terms: sorbent, regeneration, mercury, acid, water, solvent, solution, liquid, reactivation

PATENT COOPERATION TREATY

From the INTERNATIONAL BUREAU

PCT

NOTIFICATION CONCERNING TRANSMITTAL OF COPY OF INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY (CHAPTER I OF THE PATENT COOPERATION TREATY)
(PCT Rule 44bis.1(c))

To:

MADDEN, Robert B.
Schwegman, Lundberg & Woessner
P.O. Box 2938
Minneapolis, Minnesota 55402
ETATS-UNIS D'AMERIQUE

SEP 21 2015

Date of mailing *(day/month/year)*	
17 September 2015 (17.09.2015)	
Applicant's or agent's file reference	IMPORTANT NOTICE
3338021WO1	

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/US2014/019916	03 March 2014 (03.03.2014)	06 March 2013 (06.03.2013)

Applicant
ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION

The International Bureau transmits herewith a copy of the international preliminary report on patentability (Chapter I of the Patent Cooperation Treaty)

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Authorized officer Philippe Bécamel
Facsimile No. +41 22 338 82 70	e-mail: pt03.pct@wipo.int

Form PCT/IB/326 (January 2004)

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter I of the Patent Cooperation Treaty)

(PCT Rule 44*bis*)

Applicant's or agent's file reference 3338021WO1	**FOR FURTHER ACTION**	See item 4 below
International application No. PCT/US2014/019916	International filing date *(day/month/year)* 03 March 2014 (03.03.2014)	Priority date *(day/month/year)* 06 March 2013 (06.03.2013)
International Patent Classification (8th edition unless older edition indicated) See relevant information in Form PCT/ISA/237		
Applicant ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION		

1. This international preliminary report on patentability (Chapter I) is issued by the International Bureau on behalf of the International Searching Authority under Rule 44 *bis*.1(a).

2. This REPORT consists of a total of 13 sheets, including this cover sheet.

   In the attached sheets, any reference to the written opinion of the International Searching Authority should be read as a reference to the international preliminary report on patentability (Chapter I) instead.

3. This report contains indications relating to the following items:

☒	Box No. I	Basis of the report
☐	Box No. II	Priority
☒	Box No. III	Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
☒	Box No. IV	Lack of unity of invention
☒	Box No. V	Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
☐	Box No. VI	Certain documents cited
☒	Box No. VII	Certain defects in the international application
☒	Box No. VIII	Certain observations on the international application

4. The International Bureau will communicate this report to designated Offices in accordance with Rules 44*bis*.3(c) and 93*bis*.1 but not, except where the applicant makes an express request under Article 23(2), before the expiration of 30 months from the priority date (Rule 44*bis* .2).

	Date of issuance of this report 08 September 2015 (08.09.2015)
The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No. +41 22 338 82 70	Authorized officer Philippe Bécamel e-mail: pt03.pct@wipo.int

Form PCT/IB/373 (January 2004)

PATENT COOPERATION TREATY

From the
INTERNATIONAL SEARCHING AUTHORITY

To:

see form PCT/ISA/220

PCT

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY
(PCT Rule 43*bis*.1)

Date of mailing
(day/month/year) see form PCT/ISA/210 (second sheet)

Applicant's or agent's file reference
see form PCT/ISA/220

FOR FURTHER ACTION
See paragraph 2 below

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/US2014/019916	03.03.2014	06.03.2013

International Patent Classification (IPC) or both national classification and IPC
INV. B01J20/20 B01D53/02 B01D53/10 B01J20/32 C01B31/08

Applicant
ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION

1. This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☐ Box No. II Priority
- ☒ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☒ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Rule 43*bis*.1(a)(i) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☒ Box No. VII Certain defects in the international application
- ☒ Box No. VIII Certain observations on the international application

2. **FURTHER ACTION**

If a demand for international preliminary examination is made, this opinion will usually be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA") except that this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 66.1*bis*(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of 3 months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

Name and mailing address of the ISA:

European Patent Office
D-80298 Munich
Tel. +49 89 2399 - 0
Fax: +49 89 2399 - 4465

Date of completion of this opinion
see form PCT/ISA/210

Authorized Officer
Pöhlmann, Robert
Telephone No. +49 89 2399-2059

Box No. I Basis of the opinion

1. With regard to the **language**, this opinion has been established on the basis of:

   ☒ the international application in the language in which it was filed

   ☐ a translation of the international application into , which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1 (b)).

2. ☐ This opinion has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule 43bis.1(a))

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, this opinion has been established on the basis of a sequence listing filed or furnished:

   a. (means)

      ☐ on paper

      ☐ in electronic form

   b. (time)

      ☐ in the international application as filed

      ☐ together with the international application in electronic form

      ☐ subsequently to this Authority for the purposes of search

4. ☐ In addition, in the case that more than one version or copy of a sequence listing has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.

5. Additional comments:

Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non obvious), or to be industrially applicable have not been examined in respect of

☐ the entire international application

☒ claims Nos. 50-67(completely); 1-49(partially)

because:

☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international search *(specify)*:

☐ the description, claims or drawings *(indicate particular elements below)* or said claims Nos. are so unclear that no meaningful opinion could be formed *(specify)*:

☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed *(specify)*:

☒ no international search report has been established for the whole application or for said claims Nos. 50-67(completely); 1-49(partially)

☐ a meaningful opinion could not be formed without the sequence listing; the applicant did not, within the prescribed time limit:

- ☐ furnish a sequence listing on paper complying with the standard provided for in Annex C of the Administrative Instructions, and such listing was not available to the International Searching Authority in a form and manner acceptable to it.
- ☐ furnish a sequence listing in electronic form complying with the standard provided for in Annex C of the Administrative Instructions, and such listing was not available to the International Searching Authority in a form and manner acceptable to it.
- ☐ pay the required late furnishing fee for the furnishing of a sequence listing in response to an invitation under Rules 13*ter*.1(a) or (b).

☐ See Supplemental Box for further details

Box No. IV Lack of unity of invention

1. ☒ In response to the invitation (Form PCT/ISA/206) to pay additional fees, the applicant has, within the applicable time limit:

   ☐ paid additional fees

   ☐ paid additional fees under protest and, where applicable, the protest fee

   ☐ paid additional fees under protest but the applicable protest fee was not paid

   ☒ not paid additional fees

2. ☐ This Authority found that the requirement of unity of invention is not complied with and chose not to invite the applicant to pay additional fees.

3. This Authority considers that the requirement of unity of invention in accordance with Rule 13.1, 13.2 and 13.3 is

   ☐ complied with

   ☒ not complied with for the following reasons:

   see separate sheet

4. Consequently, this report has been established in respect of the following parts of the international application:

   ☐ all parts.

   ☒ the parts relating to claims Nos. 1-49(partially)

Box No. V Reasoned statement under Rule 43*bis*.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	5-49(partially)
	No:	Claims	1-4(partially)
Inventive step (IS)	Yes:	Claims	10-15, 26-37, 40-47(all partially)
	No:	Claims	1-9, 16-25, 38, 39, 48, 49(all partially)
Industrial applicability (IA)	Yes:	Claims	1-49(partially)
	No:	Claims	

2. Citations and explanations

see separate sheet

Box No. VII Certain defects in the international application

The following defects in the form or contents of the international application have been noted:

see separate sheet

Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

Re Item VIII

Certain observations on the international application

1 Clarity, Article 6 PCT)

1.1 General

The terms "about" (used in claims 4-6, 18-24, 34, 50, 58, 64), "substantially" (used in claims 5, 7-9, 50, 61-64, 67) and "core" (used in claims 3, 7, 18, 19, 51, 56, 61) are vague and unclear and leave the reader in doubt as to the meaning of the technical features to which they refer, thereby rendering the definition of the subject-matter of said claims unclear, Art. 6 PCT.

1.2 Claim 4

Claim 4 does not meet the requirements of Article 6 PCT because the matter for which protection is sought is not clearly defined. The claim attempts to define the subject-matter in terms of the result to be achieved (combines with at least 70 wt%), which merely amounts to a statement of the underlying problem, without providing the technical features necessary for achieving this result (like but not limited to volume flows, distribution of sorbent, injection of sorbent, contact times, temperatures, other components in the mercury-containing gas).

1.3 Claim 7

It is unclear (Art. 6 PCT) how the nitrogen can be homogeneously distributed in the core of the particles and how at the same time there can be a concentration profile from the surface to the core (when referring to claim 3).

1.4 Claims 8, 9

Claims 8 and 9 do not meet the requirements of Art. 6 PCT because the matter for which protection is sought is not clearly defined. The claims attempt to define the subject-matter in terms of the result to be achieved, which merely amounts to a statement of the underlying problem, without providing the technical features necessary for achieving this result.

1.5 Claim 18

It is unclear in claim 18 how a core of an activated carbon sorbent particle can comprise 99 wt% nitrogen (Art. 6 PCT). The core practically would consist of nitrogen only in this case.

1.6 Claim 20

It is unclear in claim 20 how a surface layer of an activated carbon sorbent particle can comprise 99 wt% nitrogen (Art. 6 PCT). The surface would consist of nitrogen only in this cas. Also it is unclear if the technical effect related to nitrogen being present at 0.001 wt% in the surface layer is still existing (Art.6 PCT).

1.7 Claim 22

The definition of the surface layer is unclear as a thickness of 99% of the radius of the particles is in fact the complete particle and not only a "surface layer" (Art. 6 PCT).

Re Item IV

2 **Lack of unity of invention**

Notwithstanding the objections under Article 6 PCT above, the application lacks unity within the meaning of Rule 13.1 PCT.

2.1 Reference is made to the following document:

D1 YUANJING ZHENG ET AL: "Review of technologies for mercury removal from flue gas from cement production processes",PROGRESS IN ENERGY AND COMBUSTION SCIENCE, ELSEVIER SCIENCE PUBLISHERS, AMSTERDAM, NL,vol. 38, no. 5, 26 April 2012 (2012-04-26), pages 599-629, XP028415255,ISSN: 0360-1285, DOI: 10.1016/J.PECS.2012.05.001[retrieved on 2012-05-25]

D2 DE 20 2012 003747 U1 (BLUECHER GMBH [DE]) 15 November 2012 (2012-11-15)

2.2 **There are several groups of claims:**

- Group 1: Claims 1-49 (partially), in particular claims 2-9, 16-25, 38, 39, 48, 49
- Group 2: Claims 1-49 (partially), in particular claims 10-15, 26-37, 40, 41, 44, 45, claims 50-67 (completely)
- Group 3: Claims 1-49 (partially), in particular claims 42, 43
- Group 4: Claims 1-49 (partially), in particular claims claims 46, 47

2.3 **The common concept linking together the groups is the subject-matter of claim 1**. This common concept is not new over D1.

2.3.1 D1 discloses a method of reducing the mercury content in a mercury-containing gas (see page 600, para. 1.), the method comprising obtaining or providing an activated carbon sorbent comprising activated carbon sorbent particles comprising nitrogen in a surface layer of the sorbent particles (see page 615, para. 4.2.5.1 and page 616, left column, "...by impregnation of nitrogen-containing polymers or pitches ..."), contacting a pollutant-containing gas with the activated carbon sorbent to form a pollutant-sorbent composition (implicitly disclosed as for a sorbent based removal process the pollutant has to be contacted with the sorbent, the pollutant thereby always forms a pollutant-sorbent composition), and separating at least some of the pollutant-sorbent composition from the pollutant-containing gas, to give a separated gas (see page 622, figure 20).

Hence, D1 discloses all the features of claim 1.

2.3.2 D1 further discloses

- claim 3: the concentration of nitrogen in the surface layer of the sorbent particles to be higher than in a core of the particles (see page 615, right column, lines 1-4, nitrogenated function introduced on carbon surface, therefrom following a lower nitrogenous concentration in the core of the particles)

- claim 5, 6: the mercury-containing gas to comprise a sulfur(VI) concentration, like SO_3 (see page 616, right column, 2nd paragraph)

2.4 **The differentiating features are**

- Group 1: applying the method for gases with a sulfur(VI) concentration of more than 3 ppm by mole
- Group 2: obtaining or providing the activated carbon sorbent comprises promoting an unpromoted carbon sorbent and/or providing or obtaining the unpromoted carbon sorbent from a carbon precursor comprising nitrogen and/or the activated carbon sorbent comprising a stabilizing agent
- Group 3: adding an alkaline component into the pollutant containing gas
- Group 4: regenerating the activated carbon sorbent

2.5 **with following resulting technical effects:**

- Group 1: no surprising and unexpected technical effect distinguishable

- Group 2: further enhancement of the mercury sorption efficiency of the activated carbon sorbent comprising nitrogen
- Group 3: reduction of sulfuric compounds in the pollutant containing gas
- Group 4: saving financial means for sorbent

2.6 **Thus the problems solved may be regarded as**

- Group 1: finding an alternative mercury containing gas for which the method can be applied.
- Group 2: how to modify the method of D1 in order to further enhance the mercury sorption efficiency of the activated carbon sorbent comprising nitrogen
- Group 3: how to modify the method of D1 in order to reduce the sulfuric compounds in the pollutant containing gas
- Group 4: how to modify the method of D1 in order to save financial means for sorbent

2.7 **The special technical features (R. 13.2 PCT) are**

- Group 1: **no** special technical feature A as the alternative gas is considered to be obvious for the skilled person.
- Group 2: differentiating feature = special technical feature B as the feature is neither disclosed nor rendered obvious by D1 and therefore is considered to make a contribution over D1.
- Group 3: differentiating feature = special technical feature C as the feature is neither disclosed nor rendered obvious by D1 and therefore is considered to make a contribution over D1.
- Group 4: **no** special technical feature D as the regeneration and reuse of the sorbent is disclosed by D1 (see page 612, left column, paragraph before 4.2.2.3).

2.8 **Groups of (alleged) inventions**

The groups of technical features do not solve any common technical problem. Thus the groups of claims are neither linked by the same nor by corresponding special technical features and define different inventions not linked by a single general inventive concept (Rule 13.1 and 13.2 PCT) and therefore **the requirement for unity of invention referred to in Rule 13 PCT is not fulfilled.**

2.9 **Consequence**

A search has been performed for the invention first mentioned in the claims, here the subject-matter of the claims forming group 1. Correspondingly, in the following this written opinion is restricted to group 1.

Group 1 : Claims 1-9, 16-25, 38, 39, 48, 49 (partially)

Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

3 Novelty Claim 1

3.1 As already shown under point 2.3.1 above the subject-matter of claim 1 is not new (Article 33(2) PCT).

4 Inventive Step Claim 1

Besides D1, which is considered to be the closest prior art to the subject-matter of claim 1, also D2 could be regarded as the closest prior art the subject-matter of claim 1.

4.1.1 D2 discloses

A method of reducing the pollutant content in a pollutant-containing gas (see §0142 and 0143), the method comprising:

obtaining or providing an activated carbon sorbent comprising activated carbon sorbent particles comprising nitrogen in a surface layer of the sorbent particles (see claim 1);

4.1.2 The subject-matter of claim 1 therefore differs from this known D2 in that

a pollutant-containing gas is contacted with the activated carbon sorbent, to form a pollutant-sorbent composition; and in that at least some of the pollutant-sorbent composition is separated from the pollutant-containing gas, to give a separated gas.

4.1.3 There is no unexpected technical effect related to that difference.

4.1.4 The problem to be solved by the present invention may therefore be regarded as how to to apply the method of D2 for reducing the pollutant content in a pollutant-containing gas.

4.1.5 The solution proposed in claim 1 of the present application cannot be considered to involve an inventive step (Article 33(3) PCT) for the following reasons:

It is common general knowledge for the skilled person to contact the sorbent with the gas to form a pollutant-sorbent composition and to separate the pollutant-sorbent composition from the gas to give a separated gas. Also, amongst many other documents he finds in D1 an indication (see cited passages) with which he would contact the sorbent with the gas, thereby forming a pollutant sorbent composition and separate this composition from the gas to give a separated gas, thereby arriving at the subject-matter of claim 1 without being inventive.

5 Dependent claims

Dependent claims 2-9, 16-25, 38, 39, 48, 49 do not contain any features which, in combination with the features of any claim to which they refer, meet the requirements of the PCT in respect of novelty and/or inventive step.

5.1 Claims 2, 3

The subject-matter of said claims is neither new (see points 2.3.1 and 2.3.2 above), nor inventive (see D2, claim 1 and §0143, mercury being an obvious toxic substance whose removal from gas streams with activated carbon is widely known in industry).

5.2 Claims 5, 6

D1 discloses a sulfur(VI) concentration in the mercury-containing gas (see 2.3.2 above) and the claimed ranges for said concentration are considered to be merely an arbitrary selection out of a variety of possible mercury containing gases without showing an unexpected technical effect. The properties of the activated carbon sorbent of D1 are considered to achieve the results claimed in claim 5 with regard to corresponding activated carbon sorbents.

5.3 Claims 16, 17, 38, 39, 48, 49

It is well-known in the art to add the sorbent into the mercury-containing gas and to remove it after a certain distance by various means like bag houses, electrostatic precipitators, cyclones or the like. As well, it is common general knowledge to contact the gas with the sorbent in a fixed bed, a dry or aqueous scrubber, a filter or a moving bed.

Claim 19, 21

D2 discloses a wide variety of possible nitrogen concentrations of the activated carbon (see claim 25), which is also considered to be applicable either to the surface and/or to the core of carbon particles. Therefore, the claimed ranges are not inventive.

Claim 24, 25

The claimed range of particle diameters is considered an arbitrary selection and activated carbon particles in this range are known to the skilled person (see for reference D2, §0106). Furthermore, every surface is considered to be continuous and therefore also a surface layer.

Re Item VII

Certain defects in the international application

6 The requirements of Rule 6.3(b) PCT (two-part form of independent claims) and Rule 5.1(a)(ii) and (iii) PCT (acknowledgement of prior art D1) are not met.

Electronic Acknowledgement Receipt

EFS ID:	33295181
Application Number:	15951970
International Application Number:	
Confirmation Number:	6410
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Customer Number:	21186
Filer:	Jordan Rivard Golomb/Jamie Johnson
Filer Authorized By:	Jordan Rivard Golomb
Attorney Docket Number:	4820.001US8
Receipt Date:	26-JUL-2018
Filing Date:	12-APR-2018
Time Stamp:	16:13:48
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	Non Patent Literature	0177_4820005us1_noar_07122018.pdf	1263995 487965c590894a0a0f966f433d91cf9cc2bd419e	no	8

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		Total Files Size (in bytes):	70754118		

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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.

Electronic Acknowledgement Receipt

EFS ID:	33295016
Application Number:	15951970
International Application Number:	
Confirmation Number:	6410
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Customer Number:	21186
Filer:	Jordan Rivard Golomb/Jamie Johnson
Filer Authorized By:	Jordan Rivard Golomb
Attorney Docket Number:	4820.001US8
Receipt Date:	26-JUL-2018
Filing Date:	12-APR-2018
Time Stamp:	16:07:28
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
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54	Non Patent Literature	0170_4820007us3_rerr_052220 18.pdf	656609 cdb3ef263b53e5b21a27e372f09e9dbfe94ba420	no	6
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Warnings:					
Information:					
		Total Files Size (in bytes):	42772619		

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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.

Electronic Acknowledgement Receipt

EFS ID:	33294871
Application Number:	15951970
International Application Number:	
Confirmation Number:	6410
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Customer Number:	21186
Filer:	Jordan Rivard Golomb/Jamie Johnson
Filer Authorized By:	Jordan Rivard Golomb
Attorney Docket Number:	4820.001US8
Receipt Date:	26-JUL-2018
Filing Date:	12-APR-2018
Time Stamp:	16:01:28
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
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8	Non Patent Literature	0064_response_03_24_08.pdf	10172100 b1317361e5bac87773ac9e7b8ffe3b86d8326951	no	29
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		Total Files Size (in bytes):	120699064		

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New Applications Under 35 U.S.C. 111

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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

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Substitute for form 1449A/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Complete if Known	
Application Number	15/951,970
Filing Date	April 12, 2018
First Named Inventor	Edwin S. Olson
Group Art Unit	1736
Examiner Name	Unknown

Sheet 1 of 19

Attorney Docket No: 4820.001US8

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	Application Number	15/951,970
	Filing Date	April 12, 2018
	First Named Inventor	Edwin S. Olson
	Group Art Unit	1736
	Examiner Name	Unknown
Sheet 2 of 19	Attorney Docket No: 4820.001US8	

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	"Application Serial No. 12/419,219, Response filed 05-20-11 to Non Final Office Action mailed 02-23-11", 14 pgs	
	"Application Serial No. 12/419,219, Response filed 07-15-10 to Non Final Office Action mailed 01-15-10", 22 pgs	
	"Application Serial No. 12/419,219, Response filed 10-27-11 to Non Final Office Action mailed 08-04-11", 12 pgs	
	"Application Serial No. 12/429,058, Advisory Action mailed 05-29-12", 3 pgs	
	"Application Serial No. 12/429,058, Ex Parte Quayle Action mailed 02-14-13", 7 pgs	
	"Application Serial No. 12/429,058, Examiner Interview Summary mailed 12-28-12", 4 pgs	
	"Application Serial No. 12/429,058, Final Office Action mailed 10-31-12", 21 pgs	
	"Application Serial No. 12/429,058, Final Office Action mailed 12-20-11", 21 pgs	
	"Application Serial No. 12/429,058, Non Final Office Action mailed 06-11-12", 18 pgs	
	"Application Serial No. 12/429,058, Non Final Office Action mailed 07-19-11", 20 pgs	
	"Application Serial No. 12/429,058, Notice of Allowance mailed 01-16-14", 5 pgs	
	"Application Serial No. 12/429,058, Notice of Allowance mailed 09-11-13", 11 pgs	
	"Application Serial No. 12/429,058, Response filed 01-31-13 to Final Office Action mailed 10-31-12", 19 pgs	
	"Application Serial No. 12/429,058, Response filed 03-07-13 to Ex Parte Quayle Action mailed 02-14-13", 7 pgs	
	"Application Serial No. 12/429,058, Response filed 05-11-12 to Final Office Action mailed 12-20-11", 11 pgs	
	"Application Serial No. 12/429,058, Response filed 09-11-12 to Non Final Office Action mailed 06-11-12", 16 pgs	

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	Application Number	15/951,970
	Filing Date	April 12, 2018
	First Named Inventor	Edwin S. Olson
	Group Art Unit	1736
	Examiner Name	Unknown
Sheet 9 of 19	Attorney Docket No: 4820.001US8	

OTHER DOCUMENTS – NON PATENT LITERATURE DOCUMENTS

Examiner Initial *	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, page(s), volume-issue number(s), publisher, city and/or country where published.	T1
	"Application Serial No. 12/429,058, Response filed 10-18-11 to Non Final Office Action mailed 07-19-11", 17 pgs	
	"Application Serial No. 13/427,665, Non Final Office Action mailed 01-04-13", 23 pgs.	
	"Application Serial No. 13/427,665, Non Final Office Action mailed 08-03-12", 12 pgs.	
	"Application Serial No. 13/427,665, Notice of Allowance mailed 04-16-13", 8 pgs	
	"Application Serial No. 13/427,665, Response filed 03-21-13 to Non Final Office Action mailed 01-04-13", 20 pgs	
	"Application Serial No. 13/427,665, Response filed 10-10-12 to Non Final Office Action mailed 08-03-12", 15 pgs	
	"Application Serial No. 13/427,685, Preliminary Amendment filed 03-22-12", 3 pgs	
	"Application Serial No. 13/453,274, Advisory Action mailed 11-24-14", 3 pgs	
	"Application Serial No. 13/453,274, Final Office Action mailed 10-16-14", 4 pgs	
	"Application Serial No. 13/453,274, Non Final Office Action mailed 05-30-14", 6 pgs.	
	"Application Serial No. 13/453,274, Notice of Allowance mailed 01-05-15", 5 pgs	
	"Application Serial No. 13/453,274, Response filed 09-02-14 to Non Final Office Action mailed 05-30-14", 13 pgs.	
	"Application Serial No. 13/453,274, Response filed 11-06-14 to Final Office Action mailed 10-16-14", 7 pgs.	
	"Application Serial No. 13/453,274, Response filed 12-09-14 to Advisory Action mailed 11-24-14", 6 pgs	
	"Application Serial No. 13/966,768, Non Final Office Action mailed 12-06-13", 7 pgs.	
	"Application Serial No. 13/966,768, Notice of Allowability mailed 08-01-14", 6 pgs	
	"Application Serial No. 13/966,768, Notice of Allowance mailed 04-18-14", 7 pgs	
	"Application Serial No. 13/966,768, Preliminary Amendment filed 08-14-13", 3 pgs.	
	"Application Serial No. 13/966,768, Response filed 03-06-14 to Non Final Office Action mailed 12-06-13", 6 pgs.	
	"Application Serial No. 13/966,768, Supplemental Preliminary Amendment filed 08-15-13", 6 pgs.	
	"Application Serial No. 14/102,896, Advisory Action mailed 12-16-15", 3 pgs	
	"Application Serial No. 14/102,896, Final Office Action mailed 08-26-15", 12 pgs	
	"Application Serial No. 14/102,896, Non Final Office Action mailed 03-23-15", 13 pgs	
	"Application Serial No. 14/102,896, Notice of Allowance mailed 06-10-16", 7 pgs	
	"Application Serial No. 14/102,896, Response filed 01-22-16 to Advisory Action mailed 12-16-15", 11 pgs	
	"Application Serial No. 14/102,896, Response filed 06-22-15 to Non Final Office Action mailed 03-23-15", 10 pgs	
	"Application Serial No. 14/102,896, Response filed 11-24-15 to Final Office Action mailed 08-26-15", 5 pgs	
	"Application Serial No. 14/195,360, Corrected Notice of Allowance mailed 02-27-17", 2 pgs	

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	"Application Serial No. 14/195,360, Corrected Notice of Allowance mailed 03-29-17", 2 pgs	
	"Application Serial No. 14/195,360, Final Office Action mailed 03-18-16", 18 pgs	
	"Application Serial No. 14/195,360, Final Office Action mailed 11-22-16", 7 pgs	
	"Application Serial No. 14/195,360, Non Final Office Action mailed 06-20-16", 11 pgs	
	"Application Serial No. 14/195,360, Non Final Office Action mailed 06-20-16", 11 pgs	
	"Application Serial No. 14/195,360, Non Final Office Action mailed 11-24-15", 19 pgs	
	"Application Serial No. 14/195,360, Notice of Allowance mailed 02-07-17", 5 pgs	
	"Application Serial No. 14/195,360, Response filed 01-18-17 to Final Office Action mailed 11-22-16", 7 pgs.	
	"Application Serial No. 14/195,360, Response filed 02-22-16 to Non Final Office Action mailed 11-24-15", 21 pgs.	
	"Application Serial No. 14/195,360, Response filed 05-17-16 to Final Office Action mailed 03-18-16", 15 pgs	
	"Application Serial No. 14/318,270, Advisory Action mailed 02-17-17", 4 pgs	
	"Application Serial No. 14/318,270, Corrected Notice of Allowance mailed 05-22-17", 2 pgs	
	"Application Serial No. 14/318,270, Final Office Action mailed 11-25-16", 25 pgs	
	"Application Serial No. 14/318,270, Non Final Office Action mailed 06-02-16", 24 pgs	
	"Application Serial No. 14/318,270, Notice of Allowance mailed 05-15-17", 8 pgs	
	"Application Serial No. 14/318,270, Response filed 01-25-17 to Final Office Action mailed 11-25-16", 14 pgs	
	"Application Serial No. 14/318,270, Response filed 02-27-17 to Final Office Action mailed 11-25-16", 15 pgs	
	"Application Serial No. 14/318,270, Response filed 03-02-16 to Restriction Requirement mailed 01-14-16", 9 pgs.	
	"Application Serial No. 14/318,270, Response filed 08-22-16 to Non Final Office Action mailed 06-02-16", 14 pgs.	
	"Application Serial No. 14/318,270, Restriction Requirement mailed 01-14-16", 8 pgs	
	"Application Serial No. 14/564,860, Final Office Action mailed 11-17-16", 4 pgs	
	"Application Serial No. 14/564,860, Non Final Office Action mailed 07-12-16", 6 pgs	
	"Application Serial No. 14/564,860, Notice of Allowance mailed 01-30-17", 6 pgs	
	"Application Serial No. 14/564,860, Preliminary Amendment filed 12-10-14", 8 pgs	
	"Application Serial No. 14/564,860, Preliminary Amendment filed 12-10-14", 6 pgs.	
	"Application Serial No. 14/564,860, Response filed 05-06-16 to Restriction Requirement mailed 03-16-16", 9 pgs.	
	"Application Serial No. 14/564,860, Response filed 10-03-16 to Non Final Office Action mailed 07-12-16", 11 pgs	
	"Application Serial No. 14/564,860, Restriction Requirement mailed 03-16-16", 5 pgs	
	"Application Serial No. 14/712,558, Advisory Action mailed 02-12-18", 8 pgs	

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	Examiner Name	Unknown
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	"Application Serial No. 14/712,558, Examiner Interview Summary mailed 03-05-18", 3 pgs	
	"Application Serial No. 14/712,558, Final Office Action mailed 12-06-17", 9 pgs	
	"Application Serial No. 14/712,558, Non Final Office Action mailed 07-02-18", 16 pgs	
	"Application Serial No. 14/712,558, Non Final Office Action mailed 07-06-17", 7 pgs	
	"Application Serial No. 14/712,558, Response filed 01-23-18 to Final Office Action mailed 12-06-17", 13 pgs	
	"Application Serial No. 14/712,558, Response filed 04-02-18 to Final Office Action mailed 12-06-17", 7 pgs.	
	"Application Serial No. 14/712,558, Response filed 10-06-17 to Non Final Office Action mailed 07-06-17", 12 pgs	
	"Application Serial No. 14/712,558, Supplemental Amendment filed 06-13-2018", 14 pgs.	
	"Application Serial No. 14/712,558, Supplemental Amendment Filed 5-15-2018", 6 pgs.	
	"Application Serial No. 14/712,558, Supplemental Preliminary Amendment filed 05-15-15", 6 pgs	
	"Application Serial No. 15//974,343 Preliminary Amendment Filed 5-15-2018", 7 pgs.	
	"Application Serial No. 15//978,760, Preliminary Amendment Filed 5-15-2018", 7 pgs.	
	"Application Serial No. 15/295,594, Preliminary Amendment filed 01-26-17", 8 pgs	
	"Application Serial No. 15/295,594, Preliminary Amendment filed 06-13-17", 7 pgs	
	"Application Serial No. 15/295,594, Response filed 07-03-18 to Restriction Requirement mailed 05-22-18", 9 pgs.	
	"Application Serial No. 15/295,594, Restriction Requirement mailed 05-22-18", 6 pgs	
	"Application Serial No. 15/295,594, Restriction Requirement mailed 06-29-18", 5 pgs	
	"Application Serial No. 15/295,594, Supplemental Amendment Filed 5-15-2018", 10 pgs.	
	"Application Serial No. 15/382,114, Preliminary Amendment filed 04-04-17", 3 pgs	
	"Application Serial No. 15/449,112, Advisory Action mailed 04-24-18", 3 pgs	
	"Application Serial No. 15/449,112, Final Office Action mailed 02-15-18", 13 pgs	
	"Application Serial No. 15/449,112, Non Final Office Action mailed 09-27-17", 17 pgs	
	"Application Serial No. 15/449,112, Notice of Allowance mailed 07-12-18", 8 pgs	
	"Application Serial No. 15/449,112, Response filed 01-16-18 to Non Final Office Action mailed 09-27-17", 16 pgs	
	"Application Serial No. 15/449,112, Response filed 02-15-18 to Final Office Action mailed 02-15-18", 13 pgs.	
	"Application Serial No. 15/449,112, Response filed 04-06-18 to Final Office Action mailed 02-15-18", 13 pgs	
	"Application Serial No. 15/452,527, Preliminary Amendment filed 03-08-17", 3 pgs.	
	"Application Serial No. 15/589,359, Final Office Action mailed 06-04-18", 13 pgs	
	"Application Serial No. 15/589,359, Non Final Office Action mailed 02-21-18", 15 pgs	
	"Application Serial No. 15/589,359, Preliminary Amendment filed 01-03-18", 6 pgs	

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	"Application Serial No. 15/589,359, Response filed 05-18-18 to Non Final Office Action mailed 02-21-18", 14 pgs.	
	"Application Serial No. 15/978,760 , Response filed 07-13-18 to Non Final Office Action mailed 06-29-18", 12 pgs	
	"Application Serial No. 15/978,760, Non Final Office Action mailed 06-29-18", 9 pgs	
	"Application Serial No. PCT/US04/12828, International Search Report mailed 10/22/2004", 2 pgs	
	"Application Serial No. PCT/US2014/019916, International Preliminary Report on Patentability mailed 09-17-15", 14 pgs	
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	"Bromine and its Compounds", Jolles, Z. E., Editor, Academic Press, Inc., New York, NY, (1966), pgs. 193 and 205	
	"Canadian Application Serial No. 2,523,132, Office Action mailed 07-18-11", 3 pgs	
	"Canadian Application Serial No. 2,523,132, Response filed 01-16-12 to Office Action mailed 7-18-11", 5 pgs	
	"Canadian Application Serial No. 2,584,327, Office Action mailed 03-03-09", 4 pgs.	
	"Canadian Application Serial No. 2,584,327, Office Action mailed 12-07-09", 2 pgs	
	"Canadian Application Serial No. 2,584,327, Response filed 03-19-10 to Office Action mailed 12-07-09", 8 pgs	
	"Canadian Application Serial No. 2,584,327, Response filed 09-02-09 to Office Action mailed 03-03-09", 21 pgs	
	"Canadian Application Serial No. 2,707,363, First Examiners Report Received 6/16/11", 2 pgs	
	"Canadian Application Serial No. 2,757,309, Office Action mailed 12-07-15", 3 pgs	
	"Canadian Application Serial No. 2,757,309, Response filed 06-06-16 to Office Action mailed 12-07-15", 13 pgs	
	"Canadian Application Serial No. 2,757,309, Voluntary Amendment filed 01-30-15", 27 pgs	
	"Canadian Application Serial No. 2,871,422, Response filed 02-26-16 to Office Action mailed 09-09-15", 13 pgs.	
	"Canadian Application Serial No. 2,871,422, Office Action mailed 04-02-15", 5 pgs	
	"Canadian Application Serial No. 2,871,422, Office Action mailed 04-24-18", 4 pgs	
	"Canadian Application Serial No. 2,871,422, Office Action mailed 08-31-17", 4 pgs	
	"Canadian Application Serial No. 2,871,422, Office Action mailed 09-09-15", 3 pgs	
	"Canadian Application Serial No. 2,871,422, Office Action mailed 12-22-16", 5 pgs	
	"Canadian Application Serial No. 2,871,422, Response filed 02-26-18 to Office Action mailed 08-31-17", w/ Amended Claims, 53 pgs	
	"Canadian Application Serial No. 2,871,422, Response filed 06-21-17 to Office Action mailed 12-22-16", 41 pgs	

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	"Canadian Application Serial No. 2,871,422, Response filed 08-28-15 to Canadian Office Action mailed 04-02-15", 22 pgs	
	"Chinese Application Serial No. 200480017704.4, Office Action mailed 12-08-11", (w/ English Translation), 6 pgs	
	"Chinese Application Serial No. 200480017704.4, Response filed 01-20-12 to Office Action mailed 12-08-12", (w/ English Translation of Amended Claims), 10 pgs.	
	"Chinese Application Serial No. 200580037037.0, Chinese Office Action, dated June 25, 2013 (and English translation)", no english translation, 46 pgs	
	"Chinese Application Serial No. 201380033231.6, Office Action mailed 09-21-15", w/ Partial English Translation, 6 pgs	
	"Chinese Application Serial No. 201380033231.6, Response filed 01-14-16 to Office Action mailed 09-21-15", w/English Claims, 7 pgs	
	"Chinese Application Serial No. 201480025701.9, Office Action mailed 01-16-18", (English Translation), 8 pgs	
	"Chinese Application Serial No. 201480025701.9, Office Action mailed 07-12-17", W/ English Translation, 8 pgs	
	"Chinese Application Serial No. 201480025701.9, Office Action mailed 07-25-16", (With English Translation), 4 pgs	
	"Chinese Application Serial No. 201480025701.9, Office Action mailed 12-23-16", W/ English Translation, 39 pgs	
	"Chinese Application Serial No. 201480025701.9, Office Action mailed 12-23-16", 24 pgs.	
	"Chinese Application Serial No. 201480025701.9, Response filed 03-27-18 to Office Action mailed 01-16-18", w/ English Claims, 20 pgs	
	"Chinese Application Serial No. 201480025701.9, Response filed 09-15-17 to Office Action mailed 07-12-17", w/ English Translation, 21 pgs	
	"DARCO® FGD- Powdered Activated Carbon", Norit Americas Inc. Datasheet No. 1100, (May 2009), 1 pg	
	"DATABASE WPI Week 197450", Thomson Scientific, London, GB; AN 1974-86199V XP002514926	
	"European Application Serial No. 05814011.2, Response filed 09-09-11 to European Search Report mailed 04-14-09 and Office Actioin mailed 11-05-09", 20 pgs.	
	"European Application Serial No. 05814011.2, Communication Pursuant to Article 94(3) EPC mailed 11-05-09", 4 pgs	
	"European Application Serial No. 05814011.2, Decision to grant mailed 06-08-12", 2 pgs	
	"European Application Serial No. 05814011.2, Extended European Search Report mailed 04-14-09", 13 pgs	
	"European Application Serial No. 05814011.2, Office Action mailed 01-25-08", 2 pgs.	
	"European Application Serial No. 05814011.2, Office Action mailed 01-31-12", 4 pgs	
	"European Application Serial No. 05814011.2, Office Action mailed 04-27-12", 6 pgs	

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	"European Application Serial No. 05814011.2, Response filed 02-28-12 to Office Action mailed 01-31-12", 30 pgs	
	"European Application Serial No. 05814011.2, Response filed 04-18-08 to Office Action mailed 01-25-08 and Third Party Observations submitted 12-04-07", 7 pgs.	
	"European Application Serial No. 05814011.2, Response filed 05-11-10 to Communication Pursuant to Article 94(3) EPC mailed 11-05-09", 11 pgs	
	"European Application Serial No. 05814011.2, Response filed 05-11-10 to Office Action mailed 11-05-09", 11 pgs.	
	"European Application Serial No. 05814011.2, Response filed 09-09-11 to Extended European Search Report mailed 04-14-09", 20 pgs	
	"European Application Serial No. 05814011.2, Third Party Observations submitted 03-19-12", 7 pgs.	
	"European Application Serial No. 05814011.2, Third Party Observations submitted 12-04-07", 5 pgs.	
	"European Application Serial No. 10767465.7, Decision to grant mailed 04-17-14", 2 pgs	
	"European Application Serial No. 10767465.7, Extended European Search Report mailed 12-06-12", 5 pgs	
	"European Application Serial No. 10767465.7, Intention to Grant mailed 03-17-14", 44 pgs	
	"European Application Serial No. 10767465.7, Intention to Grant mailed 09-30-13", 44 pgs	
	"European Application Serial No. 10767465.7, Response filed 02-03-14 to Intention to Grant mailed 09-30-13", 11 pgs	
	"European Application Serial No. 10767465.7, Response filed 06-20-13 to Extended European Search Report mailed 12-06-12", 18 pgs	
	"European Application Serial No. 11189249.3, Communication Pursuant to Article 94(3) EPC mailed 11-26-12", 4 pgs	
	"European Application Serial No. 11189249.3, Decision to Refuse mailed 12-19-13", 18 pgs	
	"European Application Serial No. 11189249.3, Extended European Search Report mailed 02-01-12", 7 pgs	
	"European Application Serial No. 11189249.3, Office Action mailed 11-04-13", 4 pgs	
	"European Application Serial No. 11189249.3, Response filed 02-04-13 to Communication Pursuant to Article 94(3) EPC mailed 11-26-12", 9 pgs	
	"European Application Serial No. 11189249.3, Response filed 08-28-12 to Extended European Search Report mailed 02-01-12", 9 pgs	
	"European Application Serial No. 11189249.3, Summons to Attend Oral Proceedings mailed 04-15-13", 7 pgs	

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	"European Application Serial No. 11189252.7, Decision to grant mailed 05-02-14", 2 pgs	
	"European Application Serial No. 11189252.7, Extended European Search Report mailed 01-23-12", 6 pgs	
	"European Application Serial No. 11189252.7, Extended European Search Report mailed 05-09-12", 12 pgs	
	"European Application Serial No. 11189252.7, Office Action mailed 10-30-13", 4 pgs	
	"European Application Serial No. 11189252.7, Office Action mailed 12-11-13", 6 pgs	
	"European Application Serial No. 11189252.7, Response filed 02-11-13 to Communication Pursuant to Article 94(3) EPC mailed 01-09-13", 5 pgs	
	"European Application Serial No. 11189252.7, Response filed 10-10-13 to Office Action mailed 04-15-13", 18 pgs	
	"European Application Serial No. 11189252.7, Response filed 11-22-12 to Extended European Search Report mailed 05-09-12", 13 pgs	
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	"European Application Serial No. 13719338.9, Notification Regarding Rule 164 and Article 94(3) EPC mailed 04-24-17", 8 pgs	
	"European Application Serial No. 13719338.9, Office Action mailed 05-12-17", 6 pgs	
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	"International Application Serial No. PCT/US2010/022807, International Preliminary Report on Patentability mailed 11-03-11", 6 pgs	
	"International Application Serial No. PCT/US2010/022807, International Search Report mailed 09-13-10", 3 pgs	

EXAMINER **DATE CONSIDERED**

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Substitute for form 1449A/PTO **INFORMATION DISCLOSURE STATEMENT BY APPLICANT** (Use as many sheets as necessary)	*Complete if Known*	
	Application Number	15/951,970
	Filing Date	April 12, 2018
	First Named Inventor	Edwin S. Olson
	Group Art Unit	1736
	Examiner Name	Unknown
Sheet 16 of 19	Attorney Docket No: 4820.001US8	

OTHER DOCUMENTS – NON PATENT LITERATURE DOCUMENTS		
Examiner Initial *	**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, page(s), volume-issue number(s), publisher, city and/or country where published.**	**T1**
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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Inventor(s):	Edwin S. Olson et al.	Examiner:	Unknown
Serial No.:	15/951,970	Group Art Unit:	1736
Filed:	April 12, 2018	Docket:	4820.001US8
Customer No.:	21186	Confirmation No.:	6410
Title:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY		

INFORMATION DISCLOSURE STATEMENT

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

In compliance with the duty imposed by 37 C.F.R. § 1.56, and in accordance with 37 C.F.R. §§ 1.97 *et. seq.*, the enclosed materials are brought to the attention of the Examiner for consideration in connection with the above-identified patent application. Applicant respectfully requests that this Information Disclosure Statement be entered and the documents listed on the attached PTO 1449 Form be considered by the Examiner and made of record. Pursuant to the provisions of MPEP 609, Applicant requests that a copy of the PTO 1449 Form, initialed as being considered by the Examiner, be returned to the Applicant with the next official communication.

Pursuant to 37 C.F.R. § 1.97(b), it is believed that no fee or statement is required with the Information Disclosure Statement. However, if an Office Action on the merits has been mailed after filing of the application or after the filing of the most recent RCE, the Commissioner is hereby authorized to charge the required fees to Deposit Account No. 19-0743 in order to have this Information Disclosure Statement considered.

Pursuant to 37 C.F.R. § 1.98(a)(2), copies of cited U.S. Patents and Published Applications, and Non-Published Applications identifiable by USPTO Serial Number, are no longer required to be provided to the Office. Applicant acknowledges the requirement to submit copies of foreign patent documents and non-patent literature in accordance with 37 C.F.R § 1.98(a)(2).

The Examiner is invited to contact the undersigned at the telephone number indicated if there are any questions regarding this communication.

Respectfully submitted,

SCHWEGMAN LUNDBERG & WOESSNER, P.A.
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(612) 349-9580

Date 7/26/2018 By /Nicholas P. Lanzatella/
Nicholas P. Lanzatella
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NPL:jj

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Inventor(s):	Edwin S. Olson et al.	Examiner:	Unknown
Serial No.:	15/951,970	Group Art Unit:	1736
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COMMUNICATION CONCERNING PRIOR OR COPENDING APPLICATION(S)

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

Pursuant to the guidance of MPEP §§ 2001.06(b) and 2004(9), Applicant would like to bring the following additional application(s) to the Examiner's attention. The identification of these applications is not intended to suggest that the subject matter claimed in any listed application is, or has been, substantially similar to any claim or claims in the present application.

Serial No./ Patent No.	Filing Date	Attorney Docket	Title
15/997,091	June 4, 2018	4820.001U10	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
11/209,163 7,435,286	August 22, 2005	4820.001US1	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
12/201,595	August 29, 2008	4820.001US2	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
12/419,219 8,168,147	April 6, 2009	4820.001US3	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
13/427,665 8,512,655	March 22, 2012	4820.001US4	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
13/966,768 8,821,819	August 14, 2013	4820.001US5	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
14/318,270 9,757,689	June 27, 2014	4820.001US6	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
14/712,558	May 14, 2015	4820.001US7	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

14/195,360 9,669,355	March 3, 2014	4820.003US1	ACTIVATED CARBON SORBENT INCLUDING NITROGEN AND METHODS OF USING THE SAME
15/974,343	May 8, 2018	4820.001US9	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
15/589,359	May 8, 2017	4820.003US2	ACTIVATED CARBON SORBENT INCLUDING NITROGEN AND METHODS OF USING THE SAME
15/382,114	December 16, 2016	4820.004US1	PROMOTED AMMONIUM SALT-PROTECTED ACTIVATED CARBON SORBENT PARTICLES FOR REMOVAL OF MERCURY FROM GAS STREAMS
15/449,112	March 3, 2017	4820.005US1	SORBENT COMPRISING CARBON AND NITROGEN AND METHODS OF USING THE SAME
12/429,058 8,652,235	April 23, 2009	4820.007US1	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
14/102,896 9,468,886	December 11, 2013	4820.007US2	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
15/295,594	October 17, 2016	4820.007US3	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
15/978,760	May 14, 2018	4820.007US4	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
15/853,029	December 22, 2017	4820.018US1	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

Respectfully submitted,

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Date 7/26/2018 By /Nicholas P. Lanzatella/
Nicholas P. Lanzatella
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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Inventor(s): Edwin S. Olson et al.
Title: SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

Docket No.:	4820.001US8	Serial No.:	15/951,970
Filed:	April 12, 2018	Due Date:	N/A
Examiner:	Unknown	Group Art Unit:	1736
Customer No.:	21186	Confirmation No.:	6410

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

We are transmitting herewith the following attached items (as indicated with an "X"):

X Communication Concerning Prior and Copending Applications (3 pgs.)
X Information Disclosure Statement (2 pgs.), Form 1449 (19 pgs.) Copies of Cited References (340).

If not provided for in a separate paper filed herewith, please consider this a PETITION FOR EXTENSION OF TIME for sufficient number of months to enter these papers and please charge any additional fees or credit overpayment to Deposit Account No. 19-0743. If applicable, any papers or fees supplied herewith are considered to be timely filed pursuant to 37 C.F.R. § 1.7(a), the response period falling on a Federal Holiday, Saturday or Sunday being extended to the next succeeding business day.

SCHWEGMAN LUNDBERG & WOESSNER, P.A.
Customer No.: 21186

By: /Nicholas P. Lanzatella/
Nicholas P. Lanzatella
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(11) (A) No. 1099490

(45) ISSUED 810421

(52) CLASS 23-356.5
C.R. CL. 183-34

(51) INT. CL.[2] B01D 53/34

(19) (CA) **CANADIAN PATENT** (12)

(54) PROCESS FOR REMOVING MERCURY FROM MERCURY VAPOUR-CONTAINING GASES

(72) de Jong, Geert J.;
Vos, Hendrik J.,
Netherlands

(73) Granted to Akzo N.V.
Netherlands

(21) APPLICATION No. 312,054

(22) FILED 780925

(30) PRIORITY DATE Netherlands (7 710632) 770929

No. OF CLAIMS 4 - NO DRAWING

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ABSTRACT OF THE DISCLOSURE

APR 21 1981

Removal of mercury from mercury vapour-containing gases by passing them continuously through a bed of active carbon on which a halogen is adsorbed having a thickness of at most 5 cm, while maintaining the chlorine content of the active carbon between 5 and 12% by weight. This is preferably effected by adding to said gases chlorine in an amount such that the carbon is caused to absorb a quantity thereof which equals the quantity that is consumed during the adsorption of mercury.

The present invention relates to a process for removing mercury from mercury vapour-containing gases by contacting these gases with activated carbon on which a halogen is adsorbed.

A process of the type indicated above disclosed, inter alia, in United States Patent Specifications Nos. 1,984,164 and 3,849,267. According to the former patent specification a halogen, more particularly iodine, is previously applied to the carbon, whereas according to the latter patent specification a gas containing mercury and chlorine is passed over activated carbon, as a result of which the mercury content of the gas can be reduced to 0.01 mg/m^3.

Moreover, in Soviety Chemical Industry 1972 (no. 8) p. 535-536, and from Tsvet. Metally, 1958 (31) p. 31-37, the use is disclosed of carbon loaded with chlorine as adsorbent of mercury-containing gases. In these publications mention is made of chlorine contents of the carbon which do not exceed 4% of the carbon weight. For practical purposes the methods described are not attractive in that the filter thicknesses required are unacceptably high or the residual concentrations of mercury are higher than the ones obtained with the present invention.

Applicant has found that the long-term effectiveness of these methods is very much dependent on the halogen content of the activated carbon and that a process for the continuous removal of mercury vapour from large amounts of gas is only suitable to be applied in actual practice if the activated carbon contains chlorine in an amount which is between particular critical limits.

According to the present invention in a process of the type indicated heretofore the gas to be freed from mercury vapour is continuously passed through a bed of activated carbon having a maximum thickness of 5 cm, care being taken that during said passage the activated carbon has a chlorine content in the range of from 5 to 12% by weight.

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It has been found that the use of a lower chlorine content will result in the bed retaining a low percentage of mercury. A total absence of chlorine will cause this percentage to be lower than 50%; and after a few days it will drop to a few per cent. The total amount of mercury which can be retained by the carbon then will be well below 0.1% mercury, calculated on the weight of the carbon. At a chlorine content which is somewhat lower than the minimum amount required the percentage of mercury collected will initially be good (over 90% of the mercury present can be collected initially). But this percentage will rapidly decrease to below 60% and the carbon will be found to be irreversibly clogged. In other words the effectiveness of the carbon cannot be restored by a chlorinating treatment then and regeneration of the carbon by removing the mercury collected by it is not very well feasible. At a chlorine content above the above-mentioned maximum limit the carbon is found to release much chlorine during the process and moreover, especially when the air passed through contains a high percentage of moisture, the carbon will be attacked, resulting in mechanical weakening; it must therefore be sooner replaced than when the chlorine content is within the above-specified limits. The chlorine content can be kept between the afore-mentioned limits in a simple manner by adding such an amount of chlorine to the gas that the carbon will adsorb therefrom as much chlorine as will be lost from it during the adsorption of the mercury. This loss of chlorine during the adsorption of the mercury is the result on the one hand of desorption and on the other of the formation on the carbon of mercury chloride, and is moreover due to reactions between chlorine and other constituents of the air, particularly water. It has been found that a stable situation can be reached when the chlorine content of the gas is such that per atom of mercury 15 to 25 atoms of chlorine are present in it. To this continuous feeding of chlorine there may be very special advantages. Such is the case when the

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gases to be treated already contain some chlorine, which will inevitably be present in it in a very low concentration for instance in rooms in which common salt is subjected to hydrolysis. Chlorine need then be added only until the desired level has been reached.

It should be noted that according to United States Patent Specification 3,849,267 (column 2, lines 1-10) gas containing mercury and chlorine also is brought into contact with activated carbon. In that case, however, the chlorine content is not controlled; for as minimum concentration of mercury contained in the purified gas 0.01 mg per m^3 is mentioned. This points to the fact that the chlorine content of the carbon must have been beyond the limits indicated.

Another particularly attractive way of keeping the chlorine content of the activated carbon within the limits indicated consists in that the carbon is periodically contacted with a chlorine-containing gas. In this periodic chlorine make-up it must be ensured that the chlorine content of the activated carbon does not temporarily get beyond the limits indicated, in which case the above-described disadvantages may be encountered. In actual practice it is found that when the chlorine content is made up once a week very favourable results can be obtained. For in one week the collecting capacity of the carbon for mercury is reduced by as little as less than 5 per cent. By subsequently contacting the activated carbon with a chlorine-containing gas the original collecting capacity is restored. The amount of chlorine the activated carbon is made to adsorb when it is periodically treated with a chlorine-containing gas may for instance be 0.30-0.40% of the weight of the carbon.

It has been found that use of the process according to the invention may lead to very favourable results with gases having a mercury content of about 0.01-0.15 mg/m^3. According to the inven-

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tion, this content may be reduced to 0.001-0.008 mg/m^3, which is a level that is quite acceptable under all circumstances and cannot be attained with any of the known processes. At higher mercury contents of the gas to be purified it is recommended that the air should first be subjected to some other mercury-desorbing process, for instance by washing with a sodium hypochlorite solution or using the method described in United States Patent Specification 3,849,267.

Further advantages of the present method are that the desired result can be obtained with very short contacting times in the range of 25 to 100 milliseconds and that, depending on the initial mercury content and the chosen contacting time, the percentage mercury collected is between 85 and 99%. Owing to the short contacting times there is only need to use thin (and hence inexpensive) carbon beds. Excellent results are obtained with a carbon bed having a thickness of, for instance, 2 cm. With the process according to the invention it is generally possible with 1 kg of activated carbon to treat gas at a rate of 50 to 200 m^3 per hour. In the process according to the invention the effective life of the carbon is extraordinarily long. At a mercury content of the gas of 0.05 mg/m^3 and a passage of 100 m^3/h per kg of carbon the expected effective life obtained according to the present invention is over 400 days.

Should the carbon be loaded with mercury, i.e. mercury chloride, to such a degree that its effectiveness will decrease, then it can be regenerated in a simple manner for instance by washing with an aqueous liquid containing chloride ions. In a mercury electrolysis apparatus preferably use is made for this purpose of process liquids, so that the mercury compounds released upon regeneration are fed back into the process and no secondary contamination takes place during regeneration. After the greater part of the mercury present has been removed, the carbon is washed

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with water to get rid of any clogging salts. After drying and loading with chlorine the carbon is ready to beused in a new adsorption period.

Regeneration of the carbon loaded with mercury is possible in that the mercury is present on it in the form of mercury chloride which is converted into water-soluble complex compound in a chloride-containing aqueous solution.

An unduly high moisture content of the gas from which mercury is to be removed should be avoided because the effectiveness of the activated carbon is reduced then and water is collected instead of mercury. It is preferred that the relative humidity should be kept below 75%. The relative humidity can be controlled in such a way that the excess water is isolated by previous cooling or the gas is heated to a temperature such that the desired relative humidity is obtained.

In the process according to the invention use may be made of normal activated carbon.. So there is no need to employ specially prepared carbons or other costly sorbents. Preference is given to the use of extruded carbon granules having a diameter of about 1.5 mm. The process according to the invention may be applied in the removal of mercury from, for instance, hydrogen gas. The process is particularly suitable for the removal of mercury from air which is vented from plant buildings by fans. The mercury content of such air is usually low because people who have to work in it must not be exposed to high mercury concentrations. The volumes of air to be discharged then are therefore generally large.

The process according to the invention is attractive especially in that it provides an inexpensive treatment of very large volumes of air having a low mercury content. As a result, even small environmental pollution by mercury can still be further reduced. If the air to be purified is not dust-free, this does not present any difficulty because the presence of dust does not have

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any unfavourable influence on the process.

As the usual fans generally have a small head, the air to be vented by them can be passed only through filters having a low resistance. That is why the process according to the invention is so important; for the filters to be used need only have a small thickness.

The present invention is further illustrated by way of the following Examples.

Example 1

From a room in which sodium chloride is subjected to electrolysis by the amalgamation process air is withdrawn at a rate of 10.10^6 m^3 per hour by means of 25 fans.

This air contains mercury vapour in an amount of about 0.05 mg/m^3. Per fan, this air, which has a relative humidity of 50% at a temperature of 32°C, is passed through a filter bed having a surface area of 37 m^2. The air displacement per fan is 40×10^3 m/h. The bed is formed by a 1.6 cm thick layer of granules of activated carbon 1.5 mm in diameter. The pressure drop across the filters is less than 10 mm hydrostatic head. The amount of carbon is 358 kg per fan. Before use the carbon is contacted with a chlorine-containing gas until the chlorine content is 7% by weight. The air is passed through the filters at a rate of 0.6 m/sec., so that the contacting time is 27.10^{-3} sec. After the air has left the filter, its mercury content is less than 0.004 mg/cm^3, so that the effectiveness of the filter under these conditions is more than 92%. After the filter has been in use for a week, its effectiveness has decreased to 89%.

Then air containing 60 mg of chlorine per m^3 is passed through for 30 minutes and the free chlorine content of the activated carbon is restored to 7% by weight. Upon the subsequent passage of mercury-containing air the effectiveness of the mercury removal is again found to be more than 92%.

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After one year's operation of the system in the above-described way the mercury content of the carbon is 4.3% by weight. The carbon is then regenerated by washing for 10 hours with a concentrated hydrochloric acid solution. Subsequently the hydrochloric acid is removed by washing with water, after which the carbon is dried by passing air through it.

When the carbon is dry (drying time about 1 hour), it is loaded with chlorine up to the original level, after which it is ready again for further adsorption. In an experiment in which the chlorine content is made up when the effectiveness of the mercury removal is 86%, it can be restored to 92% in the above-described way. In an experiment in which the chlorine make-up is not carried out until the effectiveness of the mercury removal has decreased to 84%, it is found that it can no longer be restored to 92% by chlorine make-up. The chlorine content has then been reduced by adsorption to a level such that the decrease has become irreversible.

Example 2

From a room in which sodium chloride is subjected to electrolysis by the amalgamation process air is withdrawn in the way described in Example 1. The air is treated in the manner indicated in Example 1, except that the carbon is not periodically brought into contact with a chlorine-containing gas. In this case the chlorine content of the carbon is kept at the desired level by having the fan air continuously contain a small amount of chlorine.

The chlorine content of the air is 0.35 p.p.m.

In this way the effectiveness of the mercury removal can be kept at 92% for a year, with the mercury content of the purified air amounting to less than 0.004 mg/m^3.

When the chlorine content of the air has been reduced until the effectiveness is less than 84%, the decrease is found to be irreversible. By increasing the chlorine feed rate it can no long be restored then to the originally high level.

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THE EMBODIMENTS OF THE INVENTION IN WHICH AN EXCLUSIVE PROPERTY OR PRIVILEGE IS CLAIMED ARE DEFINED AS FOLLOWS:

1. In a process for removing mercury from a mercury vapour-containing gas by contacting the gas with activated carbon on which a halogen is adsorbed, the improvement in which the gas to be freed from mercury vapour is continuously passed through a bed of activated carbon having a maximum thickness of 5 cm, care being taken that during said passage the activated carbon has a chlorine content in the range of from 5 to 12% by weight.

2. A process according to claim 1, in which the chlorine content of the activated carbon is kept within said limits by adding such an amount of chlorine as is lost from it during the adsorption of the mercury.

3. A process according to claim 2, in which the chlorine content of the gas is such that per atom of mercury 15 to 20 atoms of chlorine are present.

4. A process according to claim 1, in which the chlorine content of the activated carbon is kept between said limits by periodically contacting the carbon with a chlorine-containing gas.

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SUBSTITUTE

REMPLACEMENT

SECTION is not Present

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(19)(CA) **APPLICATION FOR CANADIAN PATENT** (12)

(54) Method and Apparatus for Continuously Separating Mercury from Flowing Gases

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(30) (DE) P 44 22 661.6 1994/06/28

(57) 21 Claims

Notice: This application is as filed and may therefore contain an incomplete specification.

ABSTRACT OF THE DISCLOSURE.

A method and apparatus for the continuous separation of mercury from flowing gases, in particular from exhaust gases from thermal processes, e.g. melting of mercury containing glas shards from fluorescent lamps. At least one halogen, in particular iodine and/or bromine, is added to the gases when they flow into a reaction vessel. The halogen reacts with the mercury to form mercury halide, which is separated in the solid state from the gases prior to their outflow from the reaction vessel, for example by deposition on a cooled metal surface or by filtration. Excess halogen is separated by means of adsorption. A regulating device is optionally provided which readjusts the feed rate of the halogen in such a way that a predetermined small mercury concentration is set in the outflowing gases, e.g. less than 0.2 mg per m^3 of gas.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

"METHOD AND APPARATUS FOR CONTINUOUSLY SEPARATING MERCURY FROM FLOWING GASES"

Reference to related patent and application, the disclosures of which are hereby incorporated by reference:

U.S. 4,729,882, Ide et al.;

International Application PCT/EP 92/01557, published WO 93/02773, U.S. designated, Kürzinger et al.

Reference to related patents:

European 0 289 810 A1, Vogg;

German 41 40 969 C1, Körner et al.

* * * * * * *

FIELD OF THE INVENTION.

The present invention relates to an apparatus and method for continuously separating mercury from flowing gases, in particular from exhaust gases of thermal processes. Such gases arise during operation of a glass melting installation for fragmented mercury-containing lightbulbs, e.g. fluorescent lamps or metal halide lamps.

BACKGROUND.

Conventionally, some mercury separation methods employ activated charcoal for binding the mercury. For example, the activated charcoal may be in the form of filters or screens, and

possibly used for pre-cleaning (for example WO 93/02773, Kürzinger et al.). Since activated charcoal is highly flammable, its employment is limited to cleaning gases at an appropriately low temperature.

The following methods are suitable for higher gas temperatures. Lime, to which green coke has been added, is used as the reaction agent for binding mercury, see EP 0 289 810. The mercury-green coke-adsorbate being generated is separated in a precipitator and stored. A large amount of special waste is generated, which is disadvantageous for this type of method.

Another method involves bringing the mercury- containing gases into contact with precious metal halides. In such a method, the precious metals are reduced and the corresponding mercury halide is separated. However, high costs are incurred because of the use of precious metals.

Some amalgam-forming methods are known. In the simplest case, a sheet metal plate forming an amalgam is disposed in the gas flow. The relatively small surface and the resultant short service times are disadvantageous. To avoid this problem, it is proposed in the German Patent Disclosure Document DE-OS 41 40 969 to apply amalgam-forming metals, for example gold, silver, copper, tin, zinc, palladium, iridium, or rhodium, on a substrate with a large inner specific surface (up to approximately 90 m^2/g). However, the cost of these metallized substrates is high.

A method for cleaning mercury-containing gases from municipal waste incineration plants is disclosed in US Patent No. 4,729,882, Ide et al. In a first method step the mercury is converted to water-soluble mercury chloride by the addition of chlorine gas. The mercury is washed out of the gas in a subsequent wet-chemical method step by means of a washing solution. However, the high chemical aggressiveness of chlorine is disadvantageous. Furthermore, wet washing causes an additional pressure loss in the gas flow which is equivalent to an increased energy outlay.

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THE INVENTION.

It is an object of the invention to provide an efficient method and apparatus for separating mercury (Hg) from gases on a large industrial scale and so that the threshold values for mercury concentrations in exhaust air fall below those prescribed in the relevant regulations, while overcoming the previously mentioned disadvantages.

Briefly, at least one halogen X, in particular iodine and/or bromine, is admixed directly and continuously at a controllable feed rate to mercury-containing gas in order to thereby trigger a quantitative reaction of $Hg + X_2 \longrightarrow HgX_2$. By means of this reaction the concentration of elementary mercury in the gas flow is directly reduced. The mercury halide(s) HgX_2 generated in the course of the reaction is or are subsequently separated in the solid state from the gas flow, possibly together with further mercury compounds present in the gas flow.

Iodine is a preferred reactant, because it reacts well with mercury to form mercury iodide. It is furthermore advantageous in that as compared to chlorine and fluorine, iodine is clearly less chemically aggressive. In addition, mercury iodide has a lesser water solubility (approximately 0.006%) than mercury bromide (approximately 0.62%), mercury chloride (approximately 6.9%) and mercury fluoride (dissociates), as well as a relatively high evaporation temperature (approximately 354°C). Because of this, the humidity and temperature of mercury-containing exhaust gases have a lesser effect on the separation of the mercury iodide than with the other mercury halides mentioned. In spite of this, however, it is possible in principle to employ halogens other than iodine as reactants, provided a reduced efficiency of the separation is acceptable. It is also possible to use two or more different types of halogens simultaneously as reactants.

In a first embodiment of the invention, the halogen is

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continuously supplied in manually preset amounts. Optionally, the feed rate of the halogen(s) is increased to such a degree that the gas flow shows an excess of halogen. In this manner, the elemental mercury can be completely converted to the greatest extent into mercury halide(s) and separated from the gas flow. The excess halogen contained in the gas flow after the reaction is separated in a further method step.

In a second embodiment of the method, the mercury concentration is selectively and continuously measured following the reaction. The feed rate of the halogen(s) is readjusted as a function of the measured value in such a way that a preselectable threshold value of the mercury concentration occurs, particularly less than 0.2 mg mercury per m^3 of exhaust gas. In a preferred embodiment of the method, the mercury concentration is adjusted to a value which, although lying below a prescribed threshold value, lies above zero. Overdosing with halogen is prevented in this manner, even with fluctuating mercury concentrations in the inflowing gas. Accordingly, in this embodiment it is possible to omit the additional method step for separating excess halogen, which is generally required with the uncontrolled method. A further advantage is the flexible adaptation of the supplied amount of halogen to fluctuating mercury concentrations. Halogen use is reduced to a minimum as well. This embodiment is particularly efficient.

A basic apparatus which carries out the method in accordance with the invention has a reaction vessel open on both ends, wherein the mercury-containing gases flow in through its first open end and flow out through its second open end. The flow can be created by thermal convection or by means of a blower. The reaction vessel is preferably pipe-shaped and the cross section can be arbitrary, for example circular or polygonal, e.g. rectangular. The reaction vessel has a supply or feed device for supplying at least one halogen or for placing the halogen into

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the reaction vessel.

In addition, the apparatus provides a separating device, in which the mercury halide(s) can be separated from the gas flow in the solid state, possibly together with further mercury compounds present in the gas flow. In the process, the mercury halide either (a) cools to a temperature below the evaporation temperature in the separating device or (b) flows at an already appropriate temperature into the separating device. When employing two or more halogens, cooling is performed to below the lowest evaporation temperature of the mercury halides used. For example, for mercury iodide and for mercury bromide the evaporation temperatures are approximately 354°C and 319°C, respectively. Thus, if both halogens are used, the temperature must be lower than 319°C.

In a preferred form of the first embodiment, a heat exchanger is used as the separating device. The advantage here is that by this step the energy balance is improved at the same time and in this way the efficiency of the entire system increased. In conventional installations the heat exchanger is primarily used in a manner known per se for the partial return of the process heat contained in the exhaust gas. For this purpose the heat exchanger has a large effective surface cooled by a fluid. The mercury halide(s) is (are) deposited thereon. Because of this, the effective flow cross section of the heat exchanger is increasingly reduced and as a consequence the flow resistance is increased. If it exceeds a threshold value, the heat exchanger must be cleaned. The time when a change is due is determined by means of two pressure probes, one in the inflow and the other in the outflow of the heat exchanger. With the flow speed being constant, the pressure difference between the two probes, i.e., the pressure loss caused by the heat exchanger, is a direct measurement of the flow resistance.

In the second embodiment, a filter is used as the separating

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unit, for example an electrostatic filter (E-filter) or a mechanical one. The advantage of the E-filter is a reduced pressure loss and consequently a reduced additional energy outlay. However, a mechanical filter, for example a conventional fibrous or textile filter, is cheaper.

To assure intimate mixing and reaction of mercury and halogens, a minimum distance must be maintained between the region where the halogen(s) enter(s) and where the corresponding mercury halides are separated from the gas flow. In individual cases this depends on the respective flow conditions and the type of admixture of the halogen(s). In a conventional exhaust gas installation the minimum distance is six times the hydraulic diameter of the exhaust gas conduit. It may be advantageous to assist admixing by suitable elements for the generation of flow eddies disposed in the interior of the reaction vessel.

The apparatus may have a second separating device. By means of this second separating device, halogen, which may be in excess after the reaction, is separated from the gas chamber, for example by means of an adsorption agent.

Preliminary tests with iodine have shown that aluminum chips are well suited as an adsorption medium. With dry gases it was possible to achieve an iodine absorption of 60 percent by weight, and with moist gases, such as is the general case with fossil-fired thermal processes, an iodine absorption of 40 percent by weight, at least, related to the adsorption medium. In comparison therewith, a clearly lesser iodine absorption was measured in the case of zinc chips (with dry gases, only 15 percent by weight). Aluminum chips, such as occur as waste products in machining operations, for example, are suitable. However, prior to their use as an adsorption agent, it is necessary to remove adhering lubricants which are often present.

In contrast, aluminum needles have not proven themselves

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useful because of the high apparent weight. Also, the air resistance of a corresponding filter is too great and the needles tend to adhere together, particularly with moist gases.

An apparatus for realizing the method in accordance with the invention with controlled halogen metering additionally has an Hg-measuring device for measuring the concentration of elemental mercury, as well as a control and regulating unit connected with the Hg-measuring device and acting on the feed device for the halogen. The measuring sensor of the Hg-measuring device is disposed at a distance downstream from the place where the halogen is added; the distance corresponds to at least six times the hydraulic diameter of the reaction vessel. This assures that at the place of measuring, the previously metered and supplied halogen has reacted essentially completely with the mercury and that therefore the actual remaining concentration of elemental mercury is measured.

The second separating device for separating excess halogen in the controlled apparatus may be omitted. The control unit preferably is adjusted in such a way that although the remaining concentration of mercury in the exhaust gas lies below a prescribed threshold value, it still lies above zero. In this way overdosing with halogen is impossible, i.e., the outflowing gas is sufficiently free of halogen even without an appropriate separating device.

DRAWINGS:

The invention will be explained below by means of some exemplary embodiments.

Fig. 1 is a schematic flow diagram of the method for the continuous separation of mercury from flowing gases in accordance with the invention. The additional method steps required for a controlled operation (second embodiment) are those framed by dashed lines.

Fig. 2 is schematic representation of an apparatus for

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executing the directed embodiment of the method of Fig. 1.

Fig. 3 is a schematic representation of an apparatus for executing the controlled embodiment of the method of Fig.1.

Fig. 4 is a schematic fragmentary view illustrating the separating device in form of a heat exchanger.

DETAILED DESCRIPTION:

Fig. 2 shows an apparatus 8 for performing the method of the first embodiment of the invention. The apparatus uses iodine as the reactant for the mercury and comprises the following components a tube-shaped reaction vessel 2 that is open at both ends and has a circular cross section, and a feed device 3-4-5 for introducing the iodine into the reaction vessel. The feed device includes at least one reservoir 3, which contains at least one halogen such as solid iodine and which is connected to a halogen holder 4, e.g. an evaporation trough, for charging the holder 4 with the halogen. The holder is displaceable (e.g., in a direction transverse to that of the gas flow) via a displacement unit 5 driven by a motor M. The apparatus also includes within the reaction vessel 2 a separation device 6, such as a fibrous or textile filter or an electrostatic filter 6a (Fig. 3), and an adsorption agent 7.

The feed device introduces at least one halogen into the reaction vessel 2, so that a predetermined amount of the appropriate halogen vapour is admixed with the gas flow. The evaporation holder 4 is introduced into vessel 2 by driving

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the displacement unit 5, e.g. a supply belt, with the motor M. The reservoir 3 is external to the reaction vessel 2; the temperature of the halogen within the reservoir 3 may be approximately 30°C which is less than the temperature of the gas flow within the reaction vessel 2.

The reaction vessel 2 is placed into an exhaust air conduit of a glass melting installation (not shown) for mercury-containing fragments of light bulbs. Mercury-containing gases flow through it in the direction of the arrow, wherein the gases flow in through the first open end of the reaction vessel 2 and, after having passed through the adjoining reaction area 8, flow out through the second open end of the reaction vessel, essentially free of mercury.

The evaporation holder 4 of the feed device is located in the area of the first end of the reaction vessel 2 and is charged with solid iodine from the reservoir 3. The feeding rate of gaseous iodine to supply the gas flow corresponds to the evaporation rate of the iodine and can therefore be affected by the temperature of the evaporation holder 4 and by the iodine surface exposed inside the reaction vessel, i.e., by the manually preselectable length of the partial area of the evaporation holder 4 introduced into the reaction vessel 2. A heat exchanger 25 can be located upstream of the feed device if the temperature of the Hg-laden gases entering vessel 2 is excessively high.

Because of the gas flow, the individual gas components are mixed with each other relatively quickly in the adjoining reaction region 8 and the iodine combines with the

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mercury to form mercury iodide.

Within the reaction area 8, whose length L corresponds to approximately six times the diameter D of the reaction vessel 2, the reaction vessel 2 may have a simple and therefore cost-effective separation device 6 in the form of a fibrous or textile filter, which filters out the mercury iodide. The installation position of the apparatus 1 in relation to an exhaust gas conduit (not shown) and the connection position of the upstream heat exchanger 25, bring the mercury iodide flowing through the vessel 2 and then through the fibrous or textile filter 6 to a temperature below the evaporation temperature of the iodide (approximately 354°C). The downstream-connected adsorption agent 7 is made of aluminum chips at whose surfaces possible excess iodine is adsorbed.

The particular advantage of this apparatus lies in its simple and cost-efficient design. However, the evaporation rate and therefore also the metering of the iodine is possibly affected by the temperature of the gas flowing around the evaporation trough 4. In that case it is not possible to use very small metered amounts and it may be necessary to accept overdosing.

In a particularly simply designed and therefore cost-effective embodiment, the supply or feed device consists of a wide-meshed metal net. It extends preferably over the entire cross section of the reaction vessel and is partially covered with solid iodine. The gases flow around the iodine through the uncovered mesh of the metal net, heat it and generate the desired vapor pressure. This embodiment is suitable for uses which do not require the exact control of the feed rate of halogen.

Fig. 3 schematically illustrates an apparatus 9 for carrying out the method in accordance with the second embodiment of the invention. The basic design is similar to that in Fig. 2. The flow direction has again been marked by arrows. The difference with respect to the embodiment of Fig. 2 lies in a changed feed device for the iodine, an additional regulating device and the omission of an adsorption agent.

The feed device uses a thermostat- controlled reservoir 11 of solid iodine 12, a suction line 13 provided with a blowback safety valve 14 and drying cartridge 15. The suction line 13 terminates in the reservoir 11 via a controllable pressure pump 16 and a continuous pressure line 17. A supply pipe 18 connects the reservoir 11 with the interior of the reaction vessel 10. The pressure pump 16 sucks ambient air through the drying cartridge 15 and the suction line 13 and introduces it via the pressure line 17 into the reservoir 11. The reservoir 11 is located inside a thermostat-controlled housing 19 which keeps the iodine 12 at a temperature of approximately 30°C. A suitably

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high vapor pressure of the iodine in the reservoir 11 is thereby created. The compressed air flows over the surface of the iodine 12 and is enriched with iodine vapor. The iodine-air mixture subsequently reaches the reaction vessel 10 through the supply pipe 18. The feed rate of the mixture can be controlled by means of controlling the flowing volume of the compressed air as well as the temperature and therefore the vapor pressure of the iodine 12.

The regulating device essentially includes an Hg-measuring device 20 for the selective measurement of the mercury concentration and a regulating device 21 connected therewith and controlling the controllable pressure pump 16. A measuring probe 22 of the Hg-measuring device 20 is disposed inside the reaction vessel 10 downstream of the supply pipe 18 at a distance A which approximately corresponds to six times the diameter D of the reaction vessel 10. A separation device 6 such as a fibrous or textile filter 6 (Fig. 2) or an electrostatic filter 6a (Fig. 3) is placed downstream of the measuring probe 22.

Preliminary tests have shown that this embodiment can react rapidly to fluctuations in the mercury content of the gas flow, so that with a suitable setting of the control 20, 21, no iodine overdosing occurs. For this reason an adsorption agent for iodine was omitted here, in contrast to Fig. 2.

Fig. 3 also shows, highly schematically, the separating device in form of an electrostatic precipitator 6a, connected to a voltage source 6b, the other terminal of which is connected to the vessel 10.

Fig. 4 shows, highly schematically, the separating device in form of a heat exchanger 26, located in the stream of gas flow, for use if the temperature of the gas and/or of the mercury halide goes above the evaporation temperature of the halogen being used. Heat recovered can be re-used. Cooling fluid is connected to couplings 27, 28.

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Various changes and modifications may be made, and any features described herein in connection with any one of the embodiments may be used with any of the others, within the scope of the inventive concept.

Example of an operative embodiment:

Inside diameter D of the reaction vessel 2, 10: 1.10 m temperature of gases at inlet end of reaction vessel 2, 10: 350°C gas flow rate through the reaction vessel: about 4.2 m^3/sec temperature of gases in region of introduction of halogen (Fig. 2: 4; Fig. 3: 18): 150°C rate of supply of halogen vapour from conduit 18 (Fig. 3) is such that the remaining concentration of mercury will be about 10 $\mu g/m^3$ temperature of gases with mercury halides at separator 6, 6a, 26: 140°C.

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THE EMBODIMENTS OF THE INVENTION IN WHICH AN EXCLUSIVE PROPERTY OR PRIVILEGE IS CLAIMED ARE DEFINED AS FOLLOWS:

1. A method for continuously separating mercury from a gas flow, comprising the steps of:

adding a reactant to the gas flow to form a reaction product with the mercury and thereby separate the mercury from the gas flow,

wherein, in accordance with the invention,

the reactant is at least one halogen, reacting with the mercury in the gas to form a reaction product, the reaction product including at least one mercury halide;

converting the mercury halide into solid state; and

separating the mercury halide from the gas flow.

2. The method of claim 1, wherein the step of converting includes bringing the at least one mercury halide into contact with a surface having a temperature below the evaporation temperature of the at least one mercury halide so that the at least one mercury halide deposits on this surface.

3. The method of claim 1, wherein the reaction product includes a plurality of mercury halides; and

the step of converting includes cooling the mercury halides to a temperature below a temperature which is lower than the lowest one of the evaporation temperatures of the mercury halides.

4. The method of claim 1, wherein the separation of the mercury halide from the gas flow comprises electrostatic filtering.

5. The method of claim 1, wherein the separation of the mercury halide from the gas flow comprises mechanical filtering.

6. The method of claim 1, further comprising the steps of selectively measuring the mercury concentration in the gas, and

readjusting the feed rate of the halogen as a function of the measured mercury concentration in such a way that a preselectable threshold value of the mercury concentration is set; and

wherein the steps of selectively measuring and readjusting are carried out after the reaction product is formed.

7. The method of claim 6, wherein the preselectable threshold value is less than 0.2 mg of mercury per m^3 of the gas.

8. The method of claim 1, further comprising the step of separating excess halogen from the gas flow.

9. The method of claim 8, wherein the step of separating excess halogen comprises an adsorption step.

10. The method of claim 1, wherein the at least one halogen includes an element selected from the group consisting of iodine and bromine.

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11. An apparatus for continuously separating mercury from a gas flow,

carrying out the method of claim 1,

comprising

a reaction vessel (2, 10) having a first open end through which the gas flows in and a second open end through which the gas flows out,

said reaction vessel (2, 10) having a feed device (3-4, 11-17) in the vicinity of said first open end, for supplying and placing at least one halogen into the reaction vessel, and

said reaction vessel further having a separating device (6, 6a, 26) in a vicinity of said second open end for separating mercury halide formed in the reaction vessel (2, 10) from the gas flow.

12. The apparatus of claim 11, wherein the separating device includes a heat exchanger (26).

13. The apparatus of claim 11, wherein the separating device includes an electrostatic filter (6a).

14. The apparatus of claim 11, wherein the separating device includes a mechanical filter (6), optionally a fibrous or textile filter.

15. The apparatus of claim 11, wherein the reaction vessel (2, 10) is substantially tubular in shape and the separating device and the feed device are disposed at a distance (A) from each other which corresponds to at least six times the hydraulic diameter (D) of the reaction vessel (2, 10).

16. The apparatus of claim 11, wherein said reaction vessel (2, 10) has an additional separating device (7) located in the

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vicinity of said second end, said additional separating device containing an adsorption agent for separating excess halogen from the gas flow.

17. The apparatus of claim 16, wherein the adsorption agent consists essentially of aluminum chips (7) when the halogen comprises essentially iodine.

18. The apparatus of claim 11, wherein the feed device includes

at least one halogen reservoir (3) containing the halogen, and means (4, 5) for receiving the halogen from said reservoir (3) and for controllable feeding said at least one halogen into the reaction vessel (2), so that a predetermined amount of the halogen vapor emitted from said at least one halogen is admixed with the gas flow, for reaction with the mercury within said gas flow.

19. The apparatus of claim 11, wherein the feed device has at least one reservoir (11) containing the at least one halogen (12);

means (13-17) for introducing pressurized gas into the reservoir (11); and

a halogen supply conduit (18) extending from the reservoir (11) into the reaction vessel (10) to provide a predetermined amount of the respective at least one halogen for admixture with the gas flow.

20. The apparatus of claim 19, wherein the pressurized gas introduction means includes a controlled element (16);

a halogen measuring device (20, 22) measuring the concentration of elemental mercury within the gas flow;

and a control circuit (21), responsive to the measuring

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device (20, 22) as a function of the concentration of elemental mercury, and controlling said controlled element (16) to thereby control the amount of halogen admixed to the gas flow.

21. The apparatus of claim 20, wherein the mercury measuring device (20, 22) has a measuring probe located in the the gas flow downstream of the supply conduit (18) where the at least one halogen is admixed into the gas, said probe being spaced from the conduit (18) by a distance (A) of at least six times the hydraulic diameter (D) of the reaction vessel (10).

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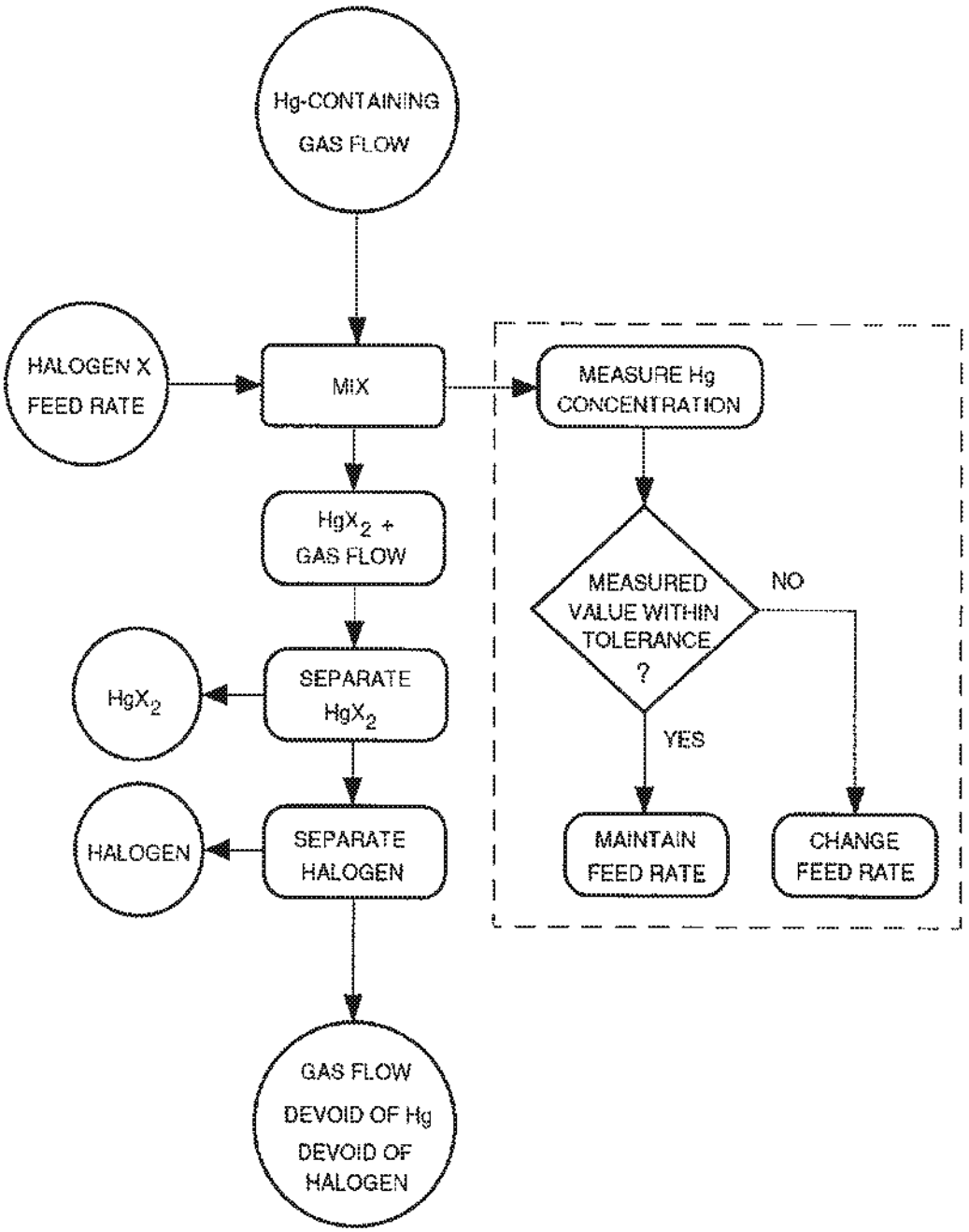


FIG. 1

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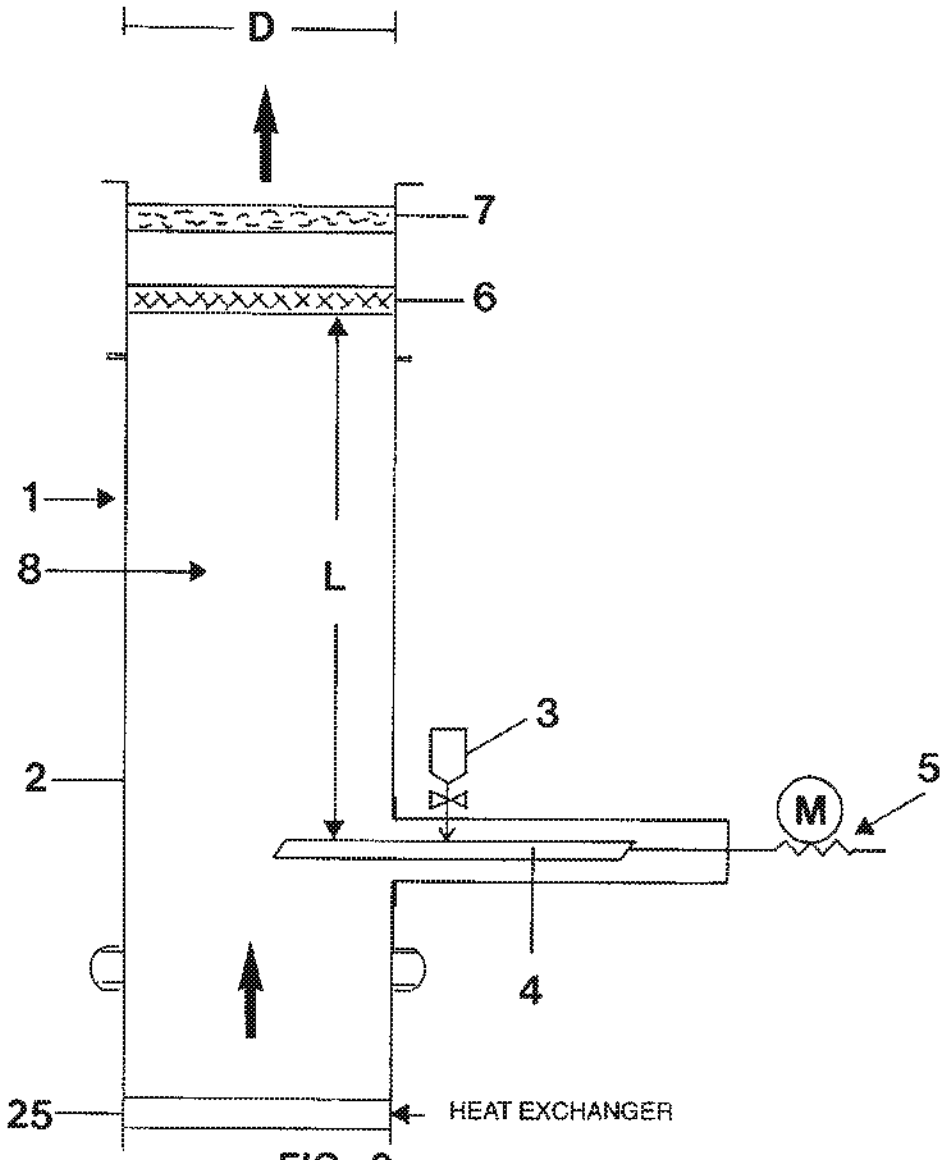


FIG. 2

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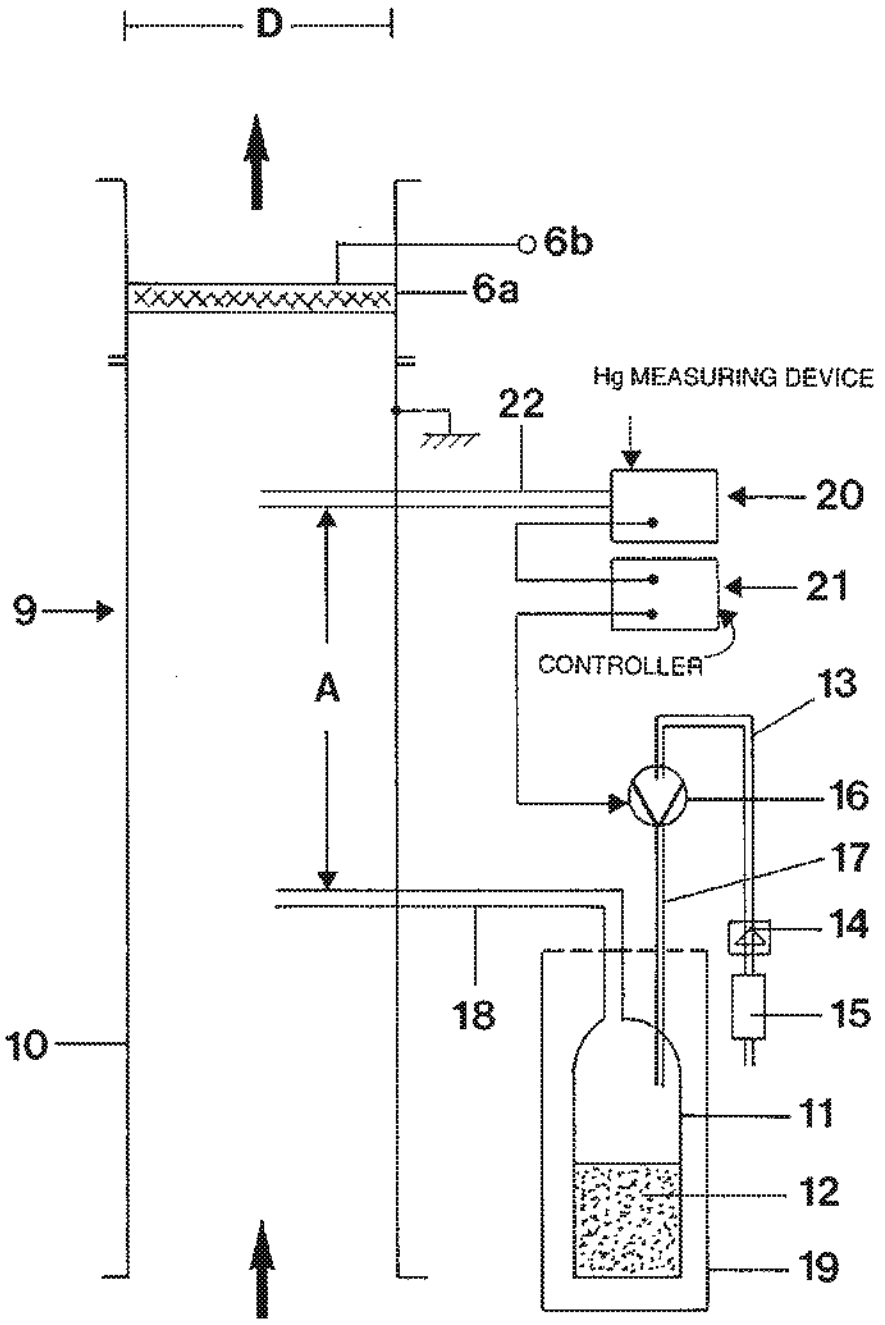


FIG. 3

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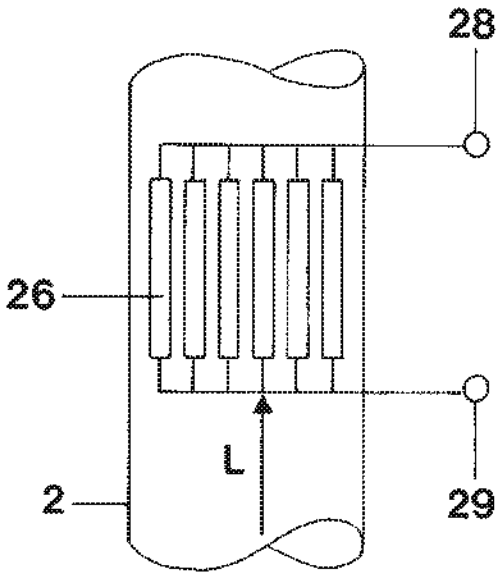


FIG. 4

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(54) METHODE DE REGENERATION DE MATERIAUX ABSORBANTS
(54) METHOD FOR THE REGENERATION OF SORBENT MATERIALS

(57)
The process of the present invention is directed to the use of sulfuric acid and/or an organic acid to regenerate spent sorbent from a metal recovery process. The sorbent can be activated carbon.

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(54) METHODE DE REGENERATION DE MATERIAUX ABSORBANTS

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(57) The process of the present invention is directed to the use of sulfuric acid and/or an organic acid to regenerate spent sorbent from a metal recovery process. The sorbent can be activated carbon.

ABSTRACT

The process of the present invention is directed to the use of sulfuric acid and/or an organic acid to regenerate spent sorbent from a metal recovery process. The sorbent can be activated carbon.

METHOD FOR THE REGENERATION OF SORBENT MATERIALS

CROSS-REFERENCE TO RELATED APPLICATION

The present application is a continuation-in-part of U.S. Patent Application entitled "Method for the Regeneration of Activated Carbon" having Serial No. 09/454,584, filed December 6, 1999, which is incorporated herein by reference.

FIELD OF THE INVENTION

The invention relates generally to the regeneration of sorbents used for metal removal and specifically to the regeneration of carbonaceous sorbents.

BACKGROUND OF THE INVENTION

Cyanidation is commonly employed for the extraction of gold from its ores. In this process, the crushed ore is treated with a dilute solution of sodium cyanide (NaCN), and a small amount of lime (CaO) to maintain a pulp pH of >9. In the presence of oxygen, gold dissolves forming gold cyanide complex.

Recovery of the gold is typically accomplished by adsorbing the gold cyanide complex on activated carbon. The gold-loaded carbon is subjected to an elution process whereby the gold is eluted from the carbon by an eluant. The eluted (spent) carbon contains a low concentration of gold and has its gold adsorption capacity reduced. The spent carbon must therefore be regenerated before reuse.

Various regeneration methods have been proposed and practiced, but all of the methods are problematical in the areas of cost, regeneration efficiency and maintenance of the mechanical strength of the regenerated carbon. Therefore, development of an effective and economical regeneration method has been eagerly desired in the art.

Some conventional methods for the regeneration of activated carbon are: (a) a low temperature heating method comprising heating spent carbon at a temperature of 100-400°C. with steam or the like, (b) a high temperature heating method comprising heating spent carbon at a temperature of 400-1000°C., (c) a regeneration method using a high temperature inert gas, (d) a chemical regeneration method using acid or alkali, and (e) a wet oxidation method, etc.

In the gold mining industry using Carbon-in-Pulp (CIP) or Carbon-in-Leach (CIL) process, good carbon activity is important. Each mining operation has to match carbon activity with process requirements. When thermal carbon regeneration is used to regenerate spent carbon, it is a general rule that the higher the carbon activity the weaker the carbon and thus higher carbon losses in these processes. Therefore, it is common practice in the industry to thermally regenerate 50% or less of the total process carbon to save on regeneration costs and to reduce carbon losses.

Chemical methods of regenerating carbon achieve partial regeneration of the carbon and can reduce the frequency of thermal regeneration. It is employed to remove specific solvents and/or compounds from the carbon. The most common of the chemical methods are; (a) acid washing the spent carbon with hydrochloric or nitric acid solutions, and (b) caustic washing with sodium hydroxide solution to remove organic contaminants.

Acid washing using nitric or hydrochloric acid at room temperature after elution may restore carbon activity to some extent, but the improved carbon activity is small compared with that of thermal regeneration.

Carbon activation with sulfuric acid by impregnation and heating at 250°C for 4-6 hours was reported to improve adsorption of ammonia by active carbon fibers.

Davidson et al., U.S. Pat. No. 4,267,069, tried to increase carbon gold adsorption capacity by contacting the carbon with an aqueous copper salt solution at a pH less than 6 in the presence of carbonic acid, carbonate or bicarbonate.

Hanceford et al., U. S. Patent No. 4,407,725, tried to regenerate activated carbon with caustic wash and an optional periodic acid wash.

Lambert et al., U. S. Patent No. 5,087,374, tried to regenerate activated carbon used to purify wastewater with an aqueous bath containing a surface-active agent while ultrasonic vibration is applied to the aqueous bath.

Activated carbon used for liquid phase, however, cannot be regenerated unless it is subjected to the high temperature heating method, namely, a re-activation or a thermal regeneration method in which regeneration is effected under the same conditions as adopted for production of activated carbon. Further, regeneration conditions, such as the amount of steam

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and the residence time in the regenerating furnace vary depending on amount and properties of activated carbons. Therefore, regeneration loss is increased by prolongation of the residence time, excessive incorporation of air and the like, and simultaneously, the mechanical strength is reduced in the regenerated activated carbon.

Furthermore, in some cases the exhaust gas from the regenerating furnace causes secondary air pollution. On the other hand, the chemical regeneration method in which high temperature heating is not conducted is advantageous in that the regeneration loss can be reduced, but the degree of regeneration is insufficient and lower than in the thermal regeneration method and both the waste coming from the spent carbon and the chemicals used for regeneration should be treated completely.

In summary, prior methods of regenerating activated carbon had disadvantages including the following:

- high energy costs of thermal regeneration;
- weakened carbon and generation of carbon fines;
- inadequate regeneration;
- expensive reagents;
- complex process; or
- expensive equipment.

Accordingly, the development of a regeneration method that will be able to overcome these defects involved in the conventional methods and to perform regeneration at an optimum regeneration rate with lessened regeneration loss has been highly demanded in the art.

As a result of intensive investigation made on regeneration of activated carbon, a novel activated carbon regeneration process has been discovered that satisfies the above-mentioned demands held in the art.

SUMMARY OF THE INVENTION

These and other needs are addressed by the process of the present invention. Generally, the present invention provides a methodology for regenerating a sorbent, such as activated carbon, for reuse in a metal adsorption, absorption, or entrapment process. As used herein, a

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"sorbent" refers to any substance that absorbs, adsorbs, or entraps another material. The methodology utilizes an organic acid and/or sulfuric acid during sorbent regeneration.

In one embodiment, the process includes the following steps:

(a) leaching (such as with a lixiviant) a metal-containing material to form a pregnant leach solution containing at least a portion of the metal,

(b) contacting the pregnant leach solution with a sorbent to form a loaded sorbent containing at least a portion of the metal dissolved in the pregnant leach solution,

(c) contacting the loaded sorbent with an eluant to form a loaded eluant containing at least a portion of the metal in or on the loaded sorbent and a spent sorbent,

(d) contacting the spent sorbent with an organic acid to form a regenerated sorbent, and

(e) recovering the metal from the loaded eluant.

The process is effective for a variety of metals and metal complexes. Preferably, the metal is a member of any one of Groups VIIIA, IB, or IIB, of the Periodic Table of the Elements. More preferably, the metal is gold, silver, platinum, copper, nickel, cobalt, mercury, and/or mixtures thereof. The metal can be complexed when in the pregnant leach solution, eluant, and/or in or on the loaded sorbent, with a variety of functional groups, including cyanide, thiocyanate, thiosulphate, and mixtures thereof. Typically, the metal is complexed with cyanide.

The lixiviant in step (a) can be any suitable material for dissolving the metal in the metal-containing material. Preferably, the lixiviant is one or more of cyanide, thiosulfate, thiourea, thiocyanate, and mixtures thereof. The leaching step can be performed by contacting the lixiviant with the material in a stirred or agitated or unstirred or unagitated vessel such as an open vessel or a closed vessel such as an autoclave or by applying the lixiviant to a pile or heap of the material or by applying the lixiviant to the material in situ.

The pregnant leach solution is contacted with the sorbent in any suitable vessel by any suitable technique. Preferably, the pregnant leach solution is contacted with the sorbent in an open or closed stirred or agitated or unstirred or unagitated vessel or by passing the pregnant leach solution through a bed of the sorbent in either a fluidized or fixed bed configuration.

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The sorbent can be any suitable material for absorbing, adsorbing, or otherwise entrapping the metal. Preferably, the sorbent is organic and more preferably the sorbent is carbon (e.g., activated carbon), resins, and/or mixtures thereof.

As in the case of the pregnant leach solution, the eluant can be contacted with the loaded sorbent in any suitable vessel by any suitable technique. Preferably, the pregnant leach solution is contacted with the sorbent in a stirred or agitated or unstirred or unagitated vessel or by passing the pregnant leach solution through a bed of the sorbent in either a fluidized or fixed bed configuration.

The eluant can include any suitable eluting materials, such as ammonia, carbohydrates (a compound having the formula $C_x(H_2O)y$ where x and y are whole numbers), inert gases such as molecular nitrogen, a metal hydroxide (a compound containing one or more OH- groups), cyanides (a compound containing one or more -CN radicals), alcohols (an organic compound containing one or more hydroxyl groups), ketones (a compound in which the carbonyl group is attached to two alkyl groups), nitriles (an organic compound containing the -C=N grouping), acetone, cyanates (a salt of cyanic acid containing the radical - CNO), hydrazines, hydroxy acids, carboxylic acids, hydrocarbons (a compound consisting only of C and H), and mixtures thereof. The eluant typically includes a solvent for the eluting material. To avoid competing with the eluting material in displacement of the metal or metal complex from the sorbent, the solvent in a preferred embodiment preferably is a polar or substantially polar compound and the eluting material is nonpolar or substantially nonpolar. Preferred solvents include water, alcohols, and mixtures thereof. The preferred elution process is described in U.S. Patent Application Serial No. 09/372,025, filed August 13, 1999, and Serial No. 09/632,386, filed August 3, 2000, both of which are incorporated herein by this reference.

The regeneration of the spent sorbent is preferably performed using an inorganic acid such as a mineral acid (*e.g.*, sulfuric acid) and/or an organic acid such as a hydroxy acid (*e.g.*, an acid containing one or more hydroxy groups such as glycolic acid and/or lactic acid) and/or a carboxylic acid (*e.g.*, an acid containing one or more carboxylic groups such as acetic acid (or methanecarboxylic acid), formic acid, caproic acid, oxalic acid, cerotic acid, aromatic acids such as benzoic acid and salicylic acid, alicyclic acids such as abietic acid and chaulmoogric acid

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and/or amino acids), with organic acids particularly hydroxy acids being preferred and hydroxycarboxylic acids (particularly glycolic acid) being even more preferred. The operating parameters for the regeneration are important to realize the optimum degree of regeneration. The pH at which the regeneration is performed typically ranges from about pH 0.5 to about pH 3 and the temperature typically from about ambient to about 95°C. The concentration of the acid in the regeneration or acid wash solution contacted with the spent sorbent preferably ranges from about 3 to about 10 % weight. Regeneration can be performed in any suitable stirred or unstirred, agitated or unagitated vessel or by passing the acid wash solution through the spent sorbent in a fluidized or fixed bed configuration with the latter being preferred.

While not wishing to be bound by any theory, it is believed that the shorter chain organic acids, particularly glycolic acid, penetrate deeply into the sorbent matrix, particularly for organic sorbents such as carbonaceous sorbents. The depth of acid penetration into the matrix is believed to be dependent upon factors such as the shape of the acid molecule and the charge distribution on the molecule. The deep penetration of the acid inhibits removal and/or neutralization of the acid when the sorbent is water rinsed (after the regenerative acid wash) to remove excess acid. Unremoved (excess) acid will consume cyanide and other basic leaching agents when the sorbent is again contacted with the pregnant leach solution. During the contacting step (b) above, the unremoved acid in the sorbent matrix is believed to cause a lower pH to exist in or near the sorbent surface and/or pores than exists in the surrounding pregnant leach solution. The lower pH causes the metal to be more strongly attracted to the sorbent, thereby causing a higher amount of metal collection in the sorbent than has been previously realized with acids such as nitric acid and hydrochloric acid, which are not believed to penetrate deeply into the sorbent matrix. Organic sorbents such as activated carbon are believed to adsorb organic acids better than inorganic acids such as nitric acid and hydrochloric acid.

In one configuration that is particularly applicable to carbon-containing sorbents, a first portion of the spent carbon is contacted with the organic acid and a second portion of the spent carbon is contacted with sulfuric acid. The first portion of the spent carbon is subsequently thermally reactivated (*e.g.*, in a kiln or other suitable reactor or heater typically at a temperature ranging from about 600 to about 800°C) before being recycled to step (b) above and the second

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portion is not subsequently thermally reactivated before being recycled to step (c) above. The presence of an excessive amount of the sulfur atom in or on the second portion may cause oxidation and loss of carbon from carbon-containing sorbents during thermal reactivation.

The regenerated sorbent will typically contain some of the acid on the surface of or inside the pores of the sorbent. For example, regenerated carbon typically includes sulfuric acid and/or the organic acid adsorbed in a plurality of pores of the carbon.

The process, in one or more of its many embodiments, can have a number of advantages. Treating the activated carbon with a solution containing organic acids, particularly glycolic acid, can restore significant gold adsorption capacity to the spent sorbent. The process can have low capital and operating costs yet high regeneration efficiency and can maintain the mechanical strength of the regenerated sorbent. Although the process uses thermal reactivation techniques in one embodiment, other embodiments employ no thermal reactivation and are conducted at ambient to below about 100° temperatures, which lowers operating (energy) costs, is energy efficient, and avoids compromising the structural integrity of the carbon (thereby generating few if any carbon fines and reducing loss of carbon). Even when thermal reactivation is employed, the spent sorbent is typically heated to relatively low temperatures, which will not seriously compromise the mechanical strength of the regenerated sorbent. The regeneration techniques of the present invention can be fast and highly effective, thereby decreasing the risk of inadequate regeneration. The process can use relatively inexpensive reagents such as sulfuric acid and glycolic acid at optimum regeneration rates. The process can be relatively simple and can use relatively inexpensive equipment. In some embodiments, the regeneration process is performed *in situ*, thereby requiring less carbon handling. The process can be readily adapted to existing plants and infrastructures. The description and drawings below show additional objects and advantages.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 shows the major components and flow directions of a typical elution process incorporating the chemical regeneration process of the eluted (spent) carbon.

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FIG. 2 shows another embodiment of a regeneration process that may be employed with any suitable elution process;

FIG. 3 shows the plot of gold adsorption rate of carbons subjected to different carbon regeneration treatment.

FIG. 4 shows carbons that have been subjected to further gold adsorption cycles to increase its gold loading capacity.

DETAILED DESCRIPTION

General Discussion

Although the process embodiments discussed below specifically refer to the elution of gold from gold-loaded activated carbon generated from an extractive metallurgical process using a cyanide-containing lixiviant and a carbohydrate-containing eluant, it is to be understood that the processes are equally applicable for metals other than gold for eluants that do not contain carbohydrates, and for processes other than extractive metallurgy. For example, the processes can be used on other precious metals and base metals, to name but a few, and may be used to elute or remove metals/metal complexes from sorbents generated in water treatment and/or purification processes.

The acid wash solution can include any suitable mineral acid, such as nitric acid, hydrochloric acid, sulfuric acid, and/or an organic acid such as glycolic acid, and preferably has a pH ranging from about pH 0.1 to about pH 5 and more preferably from about pH 0.5 to about pH 2. The concentration of the acid in the acid wash solution preferably arranges from about 0.5 to about 25 vol.% and more preferably from about 3 to about 5 vol%.

While not wishing to be bound by any theory, it is believed that the eluting or blinding agent, in the presence of the acidic pH, is at most only weakly attracted to the sorbent, thereby causing a large fraction (typically at least about 90%) of the blinding agent into the solution from the sorbent surface. In other words, in the presence of a basic pH, the equilibrium between the blinding agent in the solution and the blinding agent on the surface of the sorbent strongly favors the attachment of the blinding agent to the sorbent surface. In contrast in the presence of an

acidic pH, the equilibrium between the dissolved blinding agent and the blinding agent on the surface of the sorbent strongly favors the dissolution (or hydrolysis) of the blinding agent.

The temperature of the acid wash typically ranges from about 80 to about 120°C.

After the solvent has been contacted with the acid wash for a time typically ranging from about 1 to about 2 hours, the regenerated sorbent can be rinsed with water to remove residual acid solution.

The sorbent can then be reused in a variety of known processes to remove dissolved metal/metal complexes from pregnant lixiviant solutions, industrial affluents, waste waters, etc., to form metal/metal complex loaded sorbent for use as the feed material in any elution processes including those described above.

Preferred Embodiment -- Description

A preferred process involving selected major operations is shown in FIG. 1. The gold-loaded carbon is soaked in the carbon elution column 10 at a temperature preferably ranging from about 80 to about 100°C and more preferably about 95°C with an organic acid wash solution 14 (having about 2% w/w organic acid) typically for at least about one hour. The acid wash solution 14 is drained out of the carbon column 10 and the carbon is thereafter neutralized with a suitable base 18 such as ammonia water (having about 2% w/w base) by passing the base 18 through the column 10. The gold-loaded carbon is then eluted in the column 10 with an eluant 22 in column 10 to strip the gold from the carbon and form gold-loaded eluant 26. After the elution process, the spent carbon in column 10 is washed with hot (e.g., about 95°C) acid wash solution 30. As will be appreciated, the organic acid wash solutions 14, 30 can be stored in the same vessel or in separate vessels. Heater 34 and heat exchanger 38 are used to heat the various solutions as desired.

The reagents used in this process, e.g., glycolic acid and ammonia solutions, may be reused if necessary by draining each solution back to their respective holding tank after each batch of carbon is treated by the solution. The solution can be made up to strength with fresh reagents before reuse.

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The pre-elution treatment using organic acid solution 14 and base 18 hastens the elution process as well as helps in the regeneration of the spent carbon in the post-elution treatment with organic acid solution 30.

Preferred Embodiment -- Operation

This part describes how my invention operates in reference to FIG. 1.

As indicated in FIG. 1, the gold-loaded carbon in column 10 is prepared for elution by washing with about 1.5 to 2 Bed Volumes (BV) of about 2% weight by weight ratio (w/w) organic acid wash solution 14 at about 95°C for one hour. The organic acid solution 14 is drained and the carbon is neutralized in column 10 with about 1.5 to 2 BV of 2% w/w base or basic solution 18 for another hour. This pre-elution treatment is meant to: (a) dissolve carbonate scale and reduce levels of calcium, magnesium, aluminum, and other metals, (b) reduce overall contaminant levels in the pores of the carbon, (c) hasten the elution process, and (d) help in the chemical regeneration of the carbon.

Eluant 22 is then contacted with the loaded carbon in column 10 to form the gold-loaded eluant 26. After the elution is at least substantially completed, the eluant 22 is drained from the carbon column _10. The spent carbon is then soaked in column 10 with about 2% w/w organic acid solution 30 at about 95°C to regenerate the spent carbon. The acid wash treatment is repeated at about 1.5 to 2 BV per hour for at least about one hour. The organic acid solution 30 is then drained from column 10 for reuse.

Alternative Embodiment

In-Situ Regeneration of Carbon-- Description

An additional embodiment conducts the alternating process of metal (e.g., gold) adsorption onto the sorbent and organic acid or sulfuric acid washing on the organic sorbent (e.g., carbon) to increase its metal loading capacity.

This embodiment relates to the regeneration of the organic sorbent outside of the elution process to increase gold loading capacity.

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In-Situ Regeneration of Carbon -- Operation

In the process of recovering the metal (particularly gold) from a pregnant leach solution (particularly a cyanide solution) using an organic sorbent or sulfuric acid (particularly activated carbon), the loading capacity of the sorbent carbon can be increased by the alternating process of metal adsorption and organic acid washing.

In one configuration, the operation involves the following steps:

- Drain the gold cyanide solution from the carbon column 10 after at least a portion of the gold (or other metal) is loaded onto the carbon;
- Rinse the gold loaded carbon with water to reduce the cyanide concentrations on the carbon;
- Drain the rinse water from the column 10 and soak the carbon in the organic acid solution in column 10;
- Drain the organic acid or sulfuric acid solution from column 10 and rinse with at least about one bed volume of water to reduce acidity; and
- Put the carbon column into operation (e.g., proceed with eluting the gold from the carbon and/or regenerating the sorbent).

The process can be done in any carbon vessels and at any temperatures. The organic or sulfuric acids and basic solutions preferably use the concentrations noted above. As will be appreciated, the process can be used for other metals and other sorbents described herein.

As will be appreciated, higher acid washing temperatures will achieve faster carbon regeneration.

In another embodiment of the present invention both a first acid wash solution containing an organic acid and a second acid wash solution containing an inorganic acid or a first acid wash solution containing a first organic acid and a second acid wash solution containing a second organic acid different from the first organic acid are employed to acid wash first and second portions, respectively, of the spent sorbent. One or both of the separately washed portions can be further subjected thermal reactivation to provide a high degree of sorbent activity.

In another embodiment of the present invention, glycolic acid, sulfuric acid, or both, is used to reactivate spent sorbent that may or may not undergo thermal regeneration before reuse.

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FIG. 2 shows this process according to this embodiment when the spent sorbent is generated using a less common continuous elution process. Most spent carbon is produced batchwise using a batch elution process; therefore the alternating batches of spent sorbent are thermally reactivated or not thermally reactivated. To further save on cost, spent sorbent that will be thermally regenerated is typically not acid washed. In this configuration, only spent sorbent that will not be thermally regenerated is acid washed with glycolic or sulfuric acid by a hot, preferably acid wash.

Referring to FIG. 2, the spent sorbent 100 in the embodiment using continuous elution is first divided 104 into separate first and second portions 108, 112 by any suitable technique and in any proportion. The first portion 108 is subjected to a first acid wash 116 using a first acid wash solution 120 containing an organic acid such as glycolic acid to form an acid washed first portion 124. The acid washed first portion 124 is then thermally reactivated 128 typically at a temperature ranging from about 600 to about 800°C in a suitable reactor or heater for a time ranging from about 30 to about 60 min. to form thermally reactivated first portion 132. The second portion 112 is acid washed 136 with a second acid wash solution 140 containing an inorganic acid such as sulfuric acid to form acid washed second portion 144. The acid washed second portion 144 and thermally reactivated first portion 132 are recombined 148 by suitable techniques to form the regenerated sorbent 152, which may be reused in the sorption/elution process.

As noted, Fig. 2 reflects a process utilizing the continuous elution of spent sorbent. For a batchwise process, Fig. 2 would be modified such that alternating batches of eluted or spent sorbent would constitute the first and second portions 108, 112. The dividing step 104 and recombining step 148 would be absent from the flow schematic in this embodiment as the first and second portions would be discrete from each other. As will be appreciated, when alternating batches of spent sorbent 100 are regenerated sequentially, a separating or dividing step would not be utilized. As noted, the first portion 108 may not be acid washed before thermal reactivation to reduce costs, and the first portion 108 and second portion 112 are sequentially and alternative respectively thermally reactivated and not thermally reactivated.

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Example 1

A method for comparing carbon activity was used to compare the following carbon regeneration treatments:

- Spent carbon, with no regeneration treatment;
- Spent carbon, acid washed at 95°C with nitric acid (HN03);
- Spent carbon, acid washed at 95°"C with hydrochloric acid (HC1);
- Spent carbon, acid washed at 95°C with glycolic acid (GA); and
- Spent carbon, with thermal regeneration.

Methodology involves presoaking one gram of carbon sample in "tailings" slurry for one hour before adding the carbon to one liter of standard gold cyanide solution containing about 10 ppm gold and buffered to pH 10. The purpose of presoaking the carbon in tailings slurry was to neutralize the "pH effect" of the acid treatment on carbon activity. The carbon samples were screened to minus 8-mesh and plus 10-mesh. At 15 minutes time intervals a small aliquots of test solution were removed to determine the remaining gold concentration. A plot is prepared for each carbon sample as shown in FIG. 3.

The data shows no significant improvement in carbon activity was attributed to the use of nitric and hydrochloric acid wash. Conventional acid washing allows for the removal of minerals containing calcium, magnesium and aluminum. The removal of these minerals proves helpful in improving thermal regeneration of the carbon.

On the other hand, washing with glycolic and sulfuric acid solution, significantly improved carbon activity. It is possible that thermal regeneration can be significantly reduced or eliminated in gold-adsorption processes such as CIL or CIP.

Maximum carbon regeneration was achieved by thermal regeneration at a price of high carbon losses. It is not always necessary to have a carbon that has very high activity. The gold adsorption process will dictate the required carbon activity for optimum operation.

Example 2

FIG. 4 and Table 1 show that alternating gold adsorption and glycolic acid washing of the loaded carbon can increase the gold loading capacity of the carbon. Two spent carbons, A

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and B, each initially containing 300-ppm gold were subjected further to four periods of gold adsorption. Carbon A was not acid washed between periods while Carbon B was acid washed with glycolic acid and water rinsed between periods.

The methodology involves two grams of carbon sample added to 100 mL of 127 ppm gold solution. The gold solution contains 0.5 g/L sodium cyanide and 1 g/L calcium hydroxide. The gold adsorption period was 2 hours and the acid washing period was 15 minutes with water rinse. After each gold adsorption period the gold solution was analyzed to determine the amount of gold adsorbed by the carbon. The carbon was then put into fresh gold solution for the next gold adsorption period.

Table 1

	Gold loading (ppm gold) in the Carbon after each cycle			
Cycles	1	2	3	4
Carbon A	3820	6010	7615	9135
Carbon B	3990	7635	10785	13530

While various embodiments of the present invention have been described in detail, it is apparent that further modifications and adaptations of the invention will occur to those skilled in the art. However, it is to be expressly understood that such modifications and adaptations are within the spirit and scope of the present invention.

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What is claimed is:

1. A process for recovering a metal from a metal-containing material, comprising:

(a) leaching the material to form a pregnant leach solution containing at least a portion of the metal,

(b) contacting the pregnant leach solution with a sorbent to form a loaded sorbent containing at least a portion of the metal,

(c) contacting the loaded sorbent with an eluant to form a loaded eluant containing at least a portion of the metal in the loaded sorbent and a spent sorbent,

(d) contacting the spent sorbent with an organic acid or sulfuric acid to form a regenerated sorbent, and

(e) recovering the metal from the loaded eluant.

2. The process of Claim 1, wherein the sorbent is at least one of carbon, and resin.

3. The process of Claim 1, wherein step (a) is performed by contacting a lixiviant with the material and wherein the lixiviant includes at least of cyanide, thiosulfate, thiourea, thiocyanate and mixtures thereof.

4 The process of Claim 1, wherein the eluant includes at least one of a carbohydrate, a hydroxide, a cyanide, an alcohol, a ketone, a nitrile, acetone, a cyanate, a hydrazine, a hydroxy acid, a carboxylic acid, a hydroxycarboxylic acid, a hydrocarbon, ammonia, and mixtures thereof.

5. The process of Claim 1, wherein the metal is a member of at least one of Groups VIIIA, IB, or IIB of the Periodic Table of the Elements.

6. The process of Claim 1, wherein the organic acid is one or more hydroxy acids.

7. The process of Claim 6, wherein the hydroxy acid is one or more hydroxycarboxylic acids.

8. The process of Claim 1, wherein the regenerated sorbent is reused in step (b).

9. The regenerated sorbent made in the process of Claim 1.

10. The metal recovered in the process of Claim 1.

11. A process for treating spent carbon, comprising contacting the spent carbon with an organic acid.

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12. The process of Claim 11, further comprising:

(a) leaching the material with a lixiviant to form a pregnant leach solution containing at least a portion of the metal,

(b) contacting the pregnant leach solution with a sorbent to form a loaded sorbent containing at least a portion of the metal,

(c) contacting the loaded sorbent with an eluant to form a loaded eluant containing at least a portion of the metal in the loaded sorbent and a spent sorbent, wherein the lixiviant includes at least one of cyanide, thiosulfate, and mixtures thereof, and the eluant includes at least one of a carbohydrate, ammonia, a hydroxide, a cyanide, an alcohol, a ketone, a nitrile, acetone, a cyanate, a hydrazine, a hydroxy acid, a carboxylic acid, a hydroxycarboxylic acid, a hydrocarbon, and mixtures thereof.

13. The process of Claim 11, wherein the metal is a member of at least one of Groups VIIIA, IB, OR IIB of the Periodic Table of the Elements.

14. The process of Claim 11, wherein the organic acid is one or more hydroxy acids.

15. The process of Claim 14, wherein the hydroxy acid is one or more hydroxycarboxylic acids.

16. The process of Claim 15, wherein the hydroxycarboxylic acid is selected from the group consisting essentially of glycolic acid, lactic acid, and mixtures thereof.

17. The process of Claim 12, wherein the regenerated sorbent is reused in step (c).

18. The regenerated sorbent made in the process of Claim 11.

19. The metal recovered in the process of Claim 11.

20. The process of Claim 11, further comprising:

contacting the spent carbon with a metal-containing solution to form a metal loaded carbon;

contacting the metal loaded carbon with at least one of an organic and inorganic acid to form a treated metal loaded carbon; and

contacting the treated metal loaded carbon with an eluant.

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21. Regenerated carbon for reuse in an elution process, comprising an organic acid adsorbed in a plurality of pores of the carbon.

22. The regenerated carbon of Claim 21, wherein the organic acid is a hydroxy acid.

23. The regenerated carbon of Claim 22 wherein the hydroxycarboxylic acid is selected from the group consisting of glycolic acid, lactic acid, and mixtures thereof.

24. The regenerated carbon of Claim 22, wherein regenerated carbon includes both the sulfuric acid and the organic acid.

25. The regenerated carbon of Claim 22, wherein the regenerated carbon includes a hydroxycarboxylic acid.

26. A process for eluting a metal from a sorbent, comprising:

contacting a metal-loaded sorbent with an organic acid or sulfuric acid to form a treated metal-loaded carbon; and

contacting the treated metal-loaded carbon with an eluant to form a barren sorbent and a metal-loaded eluant containing at least most of the metal loaded onto the metal-loaded sorbent.

27. The process of Claim 26, wherein the organic acid is a hydroxy acid.

28. The process of Claim 26, wherein the hydroxy acid is selected from the group consisting of glycolic acid, lactic acid, and mixtures thereof.

29. The process of Claim 26, wherein the sorbent is at least one of activated carbon, a resin, and mixtures thereof.

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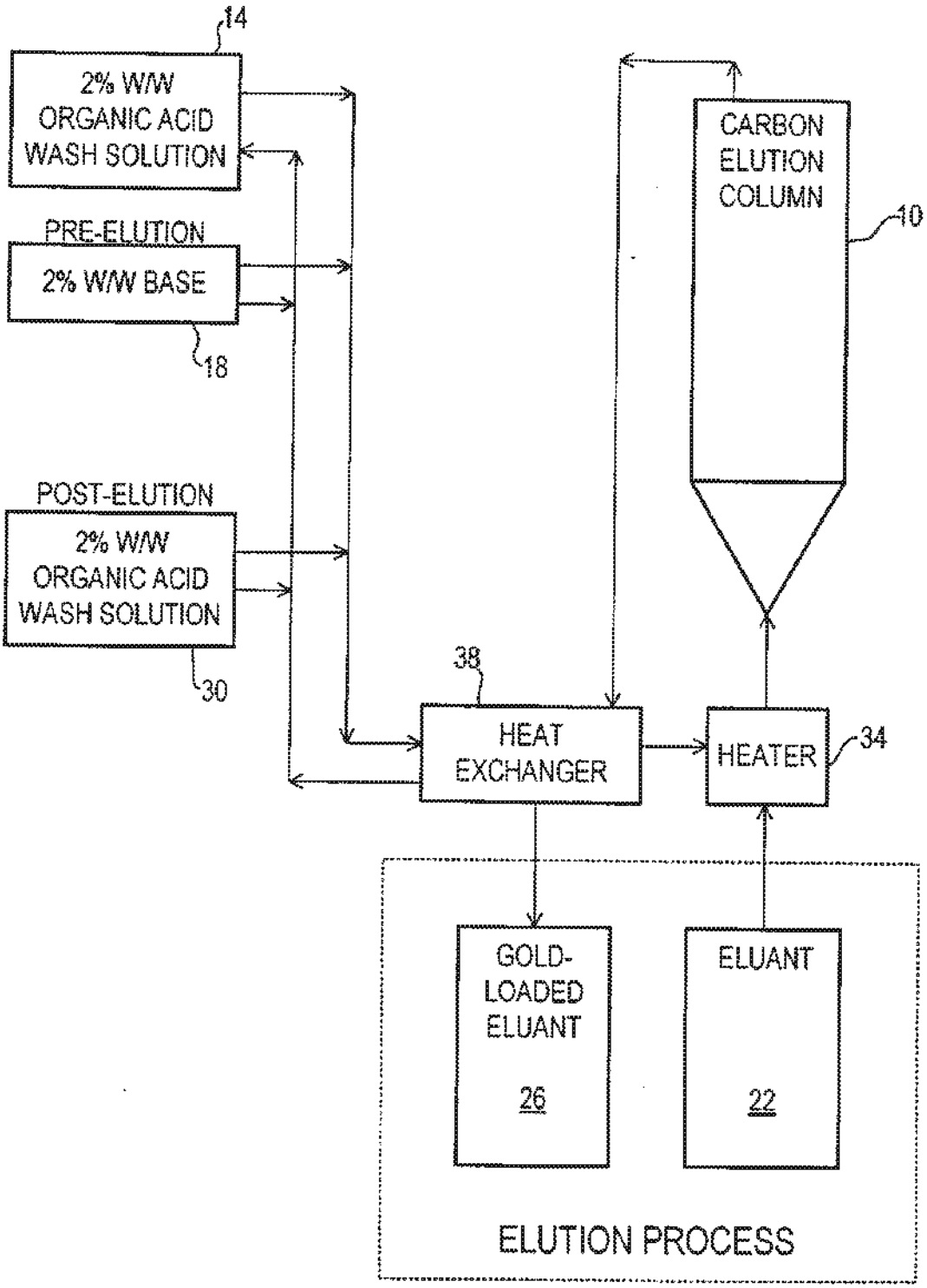

14
2% W/W
ORGANIC ACID
WASH SOLUTION
PRE-ELUTION
2% W/W BASE
18
POST-ELUTION
2% W/W
ORGANIC ACID
WASH SOLUTION
30
CARBON
ELUTION
COLUMN
10
38
HEAT
EXCHANGER
HEATER
34
GOLD-
LOADED
ELUANT
26
ELUANT
22
ELUTION PROCESS

FIG. 1

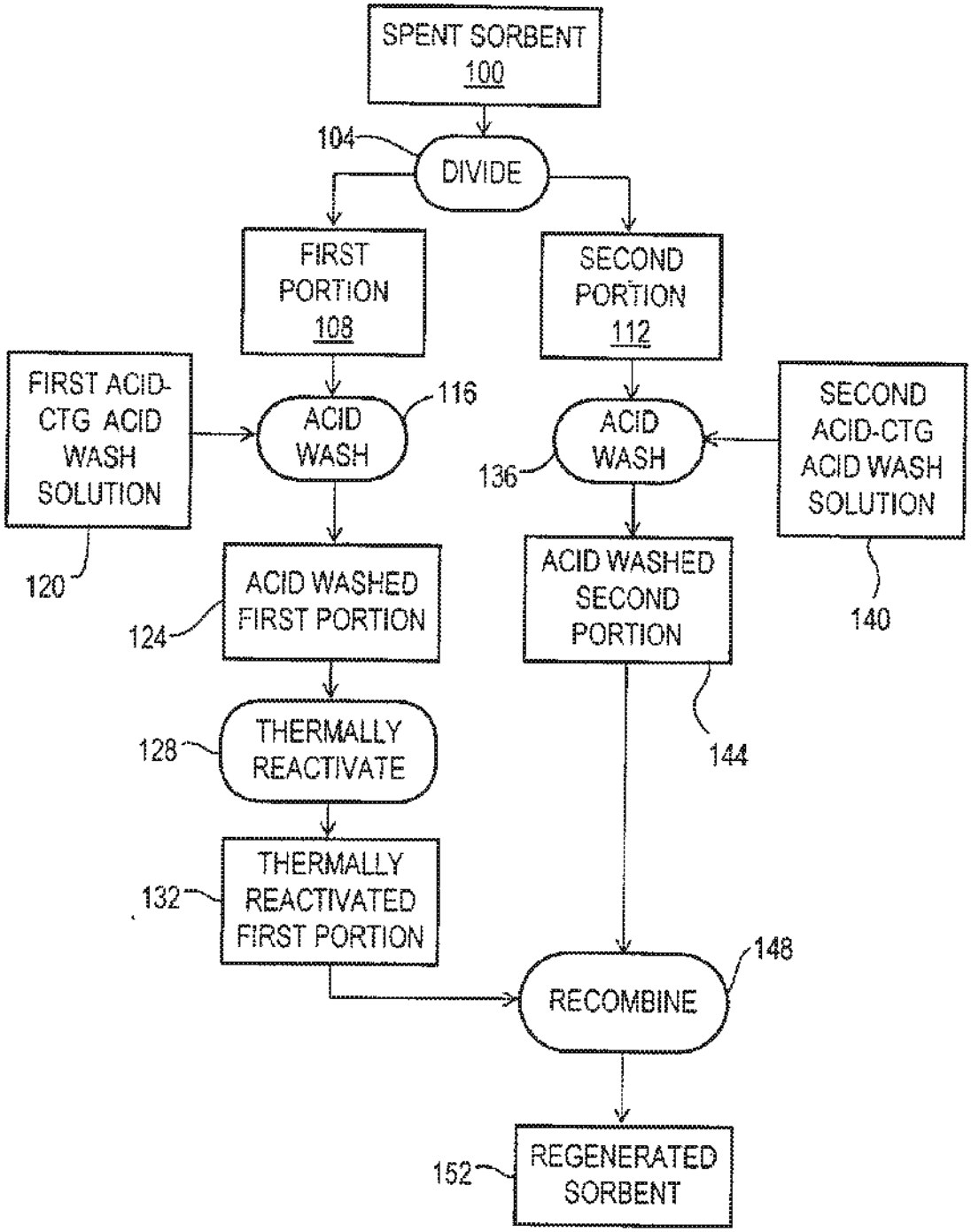

SPENT SORBENT
100
104
DIVIDE
FIRST
PORTION
108
SECOND
PORTION
112
FIRST ACID-
CTG ACID
WASH
SOLUTION
120
116
ACID
WASH
ACID
WASH
136
SECOND
ACID-CTG
ACID WASH
SOLUTION
140
ACID WASHED
FIRST PORTION
124
ACID WASHED
SECOND
PORTION
144
128
THERMALLY
REACTIVATE
132
THERMALLY
REACTIVATED
FIRST PORTION
148
RECOMBINE
152
REGENERATED
SORBENT

FIG. 2

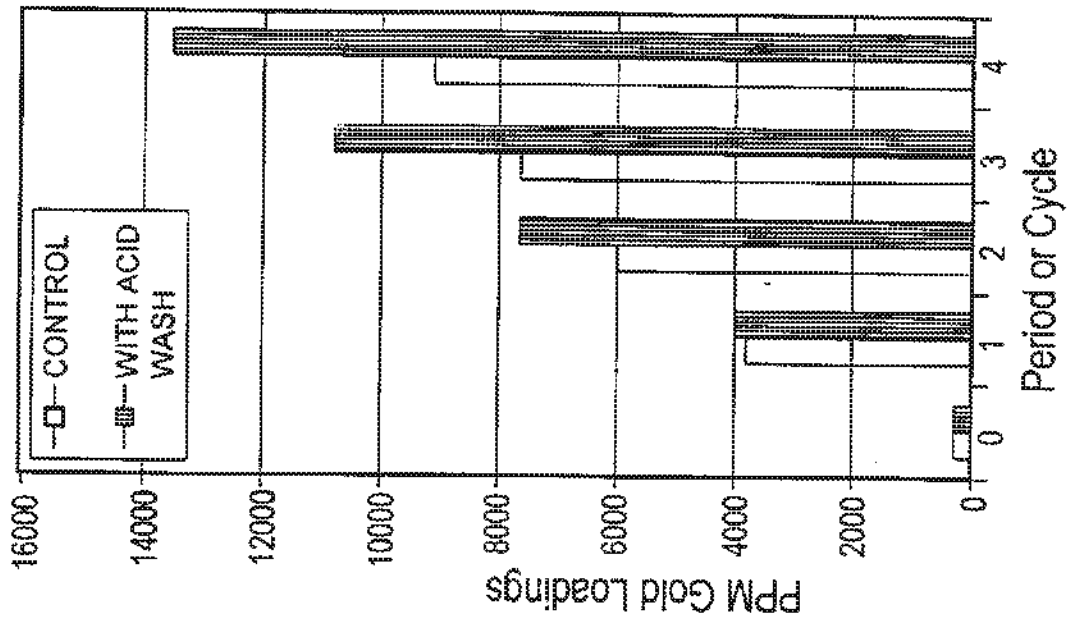

CONTROL
WITH ACID WASH
PPM Gold Loadings
Period or Cycle
16000
14000
12000
10000
8000
6000
4000
2000
0
0
1
2
3
4

FIG. 4

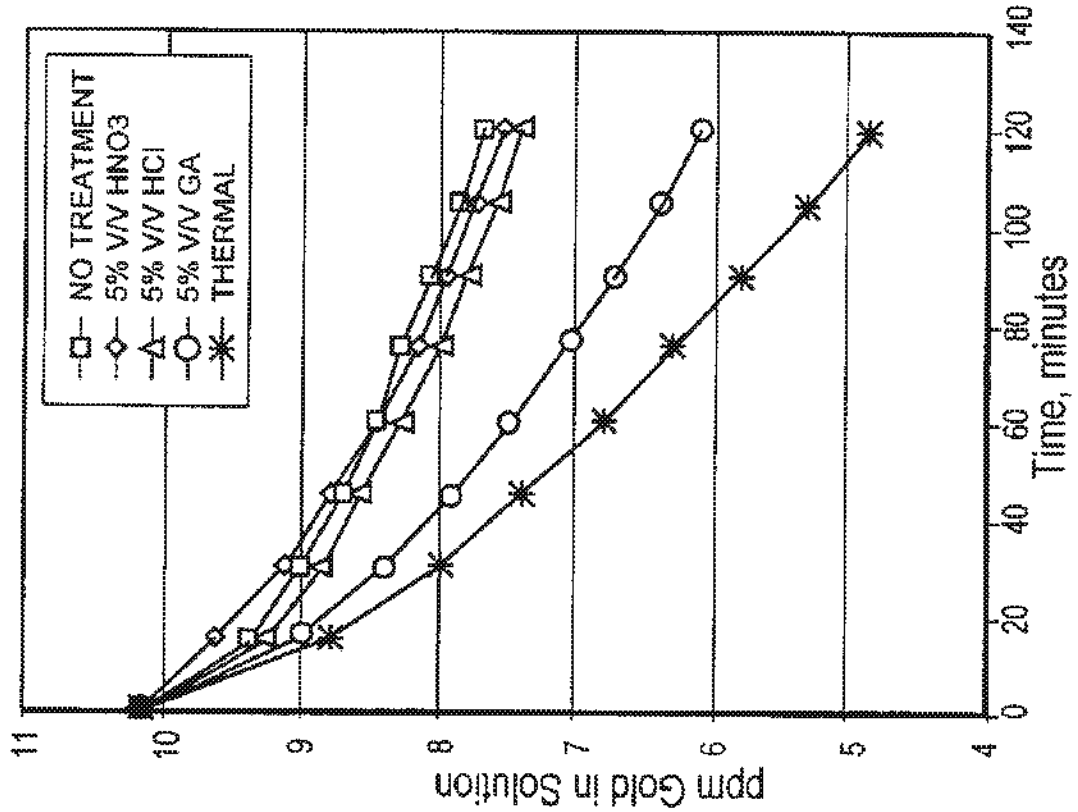

NO TREATMENT
5% V/V HNO3
5% V/V HCl
5% V/V GA
THERMAL
ppm Gold in Solution
Time, minutes
11
10
9
8
7
6
5
4
0
20
40
60
80
100
120
140

FIG. 3

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(54) POUDRE ADSORBANTE DESTINE A L'ELIMINATION DE MERCURE DANS DES COURANTS GAZEUX A TEMPERATURE ET HUMIDITE ELEVEES

(54) ADSORPTION POWDER FOR REMOVING MERCURY FROM HIGH TEMPERATURE, HIGH MOISTURE GAS STREAMS

(57)

There is disclosed an adsorption powder effective for capturing mercury from a high temperature, high moisture gas stream comprising: about 1-97% carbon; about 1-97% calcium hydroxide; about 1-97% cupric chloride impregnated carbon, and about 1-60% carbon impregnated with KI3. Additionally, a method of removing mercury from high temperature, high moisture gas streams is disclosed.

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(54) Titre : POUDRE ADSORBANTE DESTINE A L'ELIMINATION DE MERCURE DANS DES COURANTS GAZEUX A TEMPERATURE ET HUMIDITE ELEVEES

(54) Title: ADSORPTION POWDER FOR REMOVING MERCURY FROM HIGH TEMPERATURE, HIGH MOISTURE GAS STREAMS

(57) Abrégé/Abstract:
There is disclosed an adsorption powder effective for capturing mercury from a high temperature, high moisture gas stream comprising: about 1-97% carbon; about 1-97% calcium hydroxide; about 1-97% cupric chloride impregnated carbon, and about 1-60% carbon impregnated with KI3. Additionally, a method of removing mercury from high temperature, high moisture gas streams is disclosed.

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(54) Title: ADSORPTION POWDER FOR REMOVING MERCURY FROM HIGH TEMPERATURE, HIGH MOISTURE GAS STREAMS

(57) Abstract: There is disclosed an adsorption powder effective for capturing mercury from a high temperature, high moisture gas stream comprising: about 1-97% carbon, about 1-97% calcium hydroxide; about 1-97% cupric chloride impregnated carbon, and about 1-60% carbon impregnated with KI3. Additionally, a method of removing mercury from high temperature, high moisture gas streams is disclosed.

ADSORPTION POWDER FOR REMOVING MERCURY FROM HIGH TEMPERATURE, HIGH MOISTURE GAS STREAMS

BACKGROUND OF THE INVENTION

The present invention relates generally to removal of pollutants from high temperature, high moisture gas streams such as those found in devices designed to treat contaminated soil and in hazardous material incinerators. More particularly, the invention relates to the capture of mercury and other metals, dioxins, furans and other organic compounds from high temperature, high moisture gas streams using an adsorption powder containing cupric chloride.

Strict standards exist for particulate and total mercury emissions by coal-fired power plants, petroleum refineries, chemical refineries, coal fired furnaces, trash burning facilities, incinerators, metallurgical operations, thermal treatment units and other particulate and mercury emitting facilities. These same restrictions apply to mercury vapor, which can enter the atmosphere as a result of low temperature thermal desorption (LTTD) treatment of contaminated soils.

These stringent standards exist in order to protect the environment and the community. When mercury-containing gases are released, the gases disperse and mercury is deposited over a wide area. The dispersed mercury can accumulate in the soil or water supplies, where it may be incorporated into the food chain. Mercury is extremely harmful to aquatic life and ultimately to the humans who consume mercury-contaminated plants and animals. It is necessary, therefore, to have a safe and effective method of eliminating mercury from the environment.

The problem of the capture and treatment of mercury vapor, typically in the context of coal-fired power plants and waste incinerators, has been previously considered. For example, U.S. Patent No. 3,193, 987 discloses passing mercury-containing vapor over activated carbon impregnated with a metal which forms an amalgam with mercury. U.S. Patent No. 4,094,777 discloses passing a mercury-containing vapor over an adsorption mass consisting essentially of a support, sulfided

copper and sulfided silver. U.S. Patent No. 3,876,393 discloses passing mercury-containing vapors over activated carbon that has been impregnated with sulfuric acid. Selenium has also been used in the removal of mercury from a vapor. U.S. Patent No. 3,786,619 discloses passing a mercury-containing gas over a mass containing as an active component, selenium, selenium sulfide or other selenium compounds. Electrostatic precipitators and various filters have traditionally been used for mercury removal, although complex apparatus have also been disclosed. (See e.g., U.S. Patent Nos. 5,409,522 and 5,607,496.)

The problem of recapturing mercury from power plant gas streams is analogous to the need for recapturing mercury from incinerators that treat contaminated soils. A process currently in use at soil treatment facilities is known as low temperature thermal desorption (LTTD). LTTD is the main process by which contaminated soils are treated to remove mercury and other contaminants. In this process, contaminated soils are fed into a heating furnace, most commonly a rotary kiln/drum, where the soil is heated by conduction. The heating volatizes the soil components and when a thermal oxidizer is added, the components are oxidized to manageable gases, such as CO_2, Cl_2, NO_x and SO_x, where x is 1-3.

The hot gas stream is subsequently cooled. The stream may be quenched with water, which cools the stream and concurrently increases the moisture content. Although water quenching is a highly effective cooling method, this treatment increases the difficulty of removing mercury from the gas stream. The gas stream is further treated to reduce and remove metals, HCl, NO_x and SO_x using acid scrubbers, carbon beds, condensation units and through the addition of adsorption powders.

When adsorption powders are injected into the gas stream, mercury and other metals bind to moieties present in the powder, precipitating them from the gas stream. The powder-bound mercury is ultimately collected in a bag house for appropriate disposal, while the clean gas stream is exhausted to the outside atmosphere. The problem with standard LTTD methods is that some metals, such as mercury, are not removed from the stream at high efficiency and will move with

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the gas stream, ultimately into the environment. Other methods require the use of complex machinery and expensive adsorption beds. LTTD and other methods also suffer from the limitation that mercury removal from high moisture gas streams is much more difficult than mercury removal from dry streams.

Available adsorption powders remove organics, metals and other contaminants, but they do not effectively remove mercury. For example, one available powder (Sorbalite ™) consisting of carbon, calcium hydroxide and sulfur removes HCl from a gas stream, but removes only about 55-65% of the mercury. Another powder (WUELFRAsorb-C ™) consisting of alcohol saturated lime and activated carbon is also inefficient at removing mercury.

Some powders include sulfur or iodine impregnated carbon. At temperatures of 75°C or less, sulfur or iodine impregnated carbon based powders show a 95% mercury removal efficiency, however, powders formulated with sulfur impregnated carbon require a dry or low moisture gas stream to show this level of removal.

Finally, the mercury removal efficiency of the powders described and other available powders is known to be very temperature dependent, placing an additional limitation on powder formulations.

A pending application describing a process from the same inventors involved the use of solid fine cupric chloride that is not impregnated on carbon. This was found to be more difficult to handle because of the grinding process required to pulverize the cupric chloride which is generally provided in rock sizes. The grinding process requires a great amount of attention due to the dust production and associated heath hazards. Additionally, the powder is very corrosive and results in breakdown of the machinery used in the process.

Accordingly, there is a need in the industry for an adsorption powder that effectively removes metals and other organic compounds, in general, and mercury, in particular, from high temperature, high moisture gas streams generated by the incineration of contaminated soils, treatment of hazardous materials, combustion of coal and other mercury liberating sources. The powder must be inexpensive and

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easy to use. Ideally, such an adsorption powder can be employed at treatment facilities already in place and can take advantage of equipment already in position, without requiring retooling or reconfiguring existing equipment.

SUMMARY OF THE INVENTION

There is disclosed an adsorption powder and method for removing mercury and other metals and contaminants from a high temperature, high moisture gas stream comprising: about 1-97% carbon; about 1-97% calcium hydroxide; about 1-97% cupric chloride impregnated carbon and about 1-60% KI_3 impregnated carbon. A method for removing mercury and other metals, dioxins, furans and other organic compounds from high temperature, high moisture gas streams using the claimed powder is also disclosed.

BRIEF DESCRIPTION OF THE DRAWINGS

The instant invention will be more fully understood in the following detailed description, it being understood, however, that the invention is not confined to the precise disclosure. Changes and modifications may be made that do not affect the spirit of the invention, nor exceed the scope thereof, as expressed in the appended claims. Accordingly, the instant invention will now be described with particular reference to the accompanying drawings.

Figure 1 is a schematic diagram illustrating the design of an LTTD facility in which the claimed adsorption powder can be used to remove mercury from gas streams.

DETAILED DESCRIPTION OF THE INVENTION

There is disclosed an adsorption powder for removing mercury and other metals from a gas stream comprising:

a) about 1-97% carbon;

b) about 1-97% calcium hydroxide;

c) about 1-97% cupric chloride impregnated carbon; and

d) about 1-60% KI_3 impregnated carbon.

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The powder more particularly comprises about 10-70% carbon, about 20-80% calcium hydroxide, about 1-50% cupric chloride impregnated carbon and about 1-50% KI_3 impregnated carbon. A preferred adsorption powder comprises about 20-60% carbon, about 30-70% calcium hydroxide, about 1-30% cupric chloride impregnated carbon and about -30% KI_3 impregnated carbon.

In a more preferred embodiment, the adsorption powder comprises about 25-45% carbon, about 40-60% calcium hydroxide, about 1-15% cupric chloride impregnated carbon and about 1-15% KI_3 impregnated carbon. The most preferred embodiment comprises about 40% carbon, about 40% calcium hydroxide, about 10% cupric chloride impregnated carbon and about 10% KI_3 impregnated carbon.

The use of the KI_3 impregnated carbon has been found to be optional as the powder lacking this component performed equally as well as one containing the KI_3 impregnated carbon.

Thus, in another embodiment of the invention, , the adsorption powder comprises about 10-70% carbon, about 20-80% calcium hydroxide and about 1-50% cupric chloride impregnated carbon.

There is also disclosed a process for removing mercury from a gas stream using the adsorption powder of the invention comprising the steps of:

a) placing a solid phase mercury-containing contaminated soil feed into a rotary kiln/drum;

b) heating said kiln/drum containing said soil feed to form gaseous and solid components of the sample;

c) transferring the gaseous component of said soil feed to an exhaust cleaning unit/afterburner and the solid component of clean soil to a soil cooling unit;

d) heating the gaseous component of said contaminated soil feed in said exhaust cleaning unit/afterburner;

e) cooling the gaseous component of said contaminated soil feed;

f) adding the adsorption powder of Claim 1 to the gaseous component;

g) transferring the powder-containing gaseous component to a baghouse; and

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h) releasing the substantially mercury-free gaseous component of said sample to the atmosphere.

An adsorption powder for the removal of mercury and other metals, dioxins, furans and other organic compounds must be efficient under a range of conditions. Currently available powders do not function well at high temperatures and in high moisture environments, conditions that are favorable to mercury removal.

High temperatures are necessary for effective removal of contaminants from soil. Temperatures of about 1800°F are necessary to volatize organics, metals and other impurities from the contaminated soil. Mercury that is trapped in contaminated soil, however, is most efficiently adsorbed on carbon at about 300-500°F. The most practical method of cooling a gas stream exiting an 1800°F oven is to inject water into the gas stream. Water injection cools the gas stream to a temperature favorable to mercury removal, but also increases the moisture content of the sample, which decreases the efficiency of available mercury adsorption powders. The mercury absorbing properties of available powders suffer dramatically in a high moisture environment. The adsorption powder of the invention, however, operates effectively even in a higher moisture environment.

Experiments with carbon sources showed that coal carbon was superior to wood carbon for mercury adsorption. Many available adsorption powders use wood carbon as a component, rather than coal carbon. Cupric chloride was observed to significantly enhance the adsorption of mercury from a gas stream and is the key to the instant invention. Cupric chloride supplies chlorine and activated copper to the elemental mercury in the exhaust stream. Elemental mercury reacts with the chlorine to form mercury chloride and the activated copper to form a stable mercury amalgam. Both forms of mercury are easily captured from the exhaust gas stream. KI_3 impregnated carbon was also found to increase mercury adsorption when it was included in the powder.

Figure 1 shows a schematic diagram of the actual process and equipment used to carry out the invention. Prescreened contaminated feed soil ready to be processed **2** is placed within soil cleaning unit **4**. The contaminated soil is heated to about 900°F or a temperature that will completely volatize the contaminates from the soil and generate a gas stream, as well as a clean/remediated solid soil component. Preferably, soil cleaning unit **4** is a rotary kiln. The gas stream is then passed out

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of soil cleaning unit **4** to dust remover **6**, while any solid fraction of the feed soil is transferred to clean soil cooling unit **8**, where the soil is cooled and prepared for reuse. Dust remover **6** is preferably a multi-tube dust collector.

After dust remover **6** removes any particulate matter from the gas stream sample, the gas stream is passed into the Exhaust Cleaning Unit **10**. The Exhaust Cleaning Unit heats the volatilized contaminates to a temperature of about 1800°F for a minimum of two seconds retention time, which assures complete destruction of any remaining organic or other contaminants. From the Exhaust Cleaning Unit **10**, the gas stream then passes through cooling chamber **12** wherein a water pump (not shown) injects water into the cooling chamber **12** to lower the temperature of the sample to about 360°F. This cooling process consequentially increases the moisture content of the sample.

The high temperature, high moisture gas stream is then contacted to the adsorption powder of the invention, which is stored in adsorbent storage silo **14** and injected into the gas stream. This powder formulation is effective in removing metals, particularly mercury, and other contaminants.

After the gas stream has been contacted to the adsorption powder, the powder/gas stream mixture continues on to baghouse **16**. The carbon component of the adsorption powder collects on the walls of bags and acts as a particulate filter for the gases leaving the baghouse. Baghouse **16** collects the particulate mercury-containing fraction of the adsorption powder mixture, which is transported to a suitable bulk storage facility **20** and subsequently removed. The gaseous fraction is released to the outside atmosphere through vent **18**, while the remaining dust particulate fraction is handled in a similar manner to the particulate mercury fraction of the adsorption powder mixture **20**.

EXAMPLE 1

A series of field tests were performed employing a preferred adsorption powder formulation. Three soil samples containing about 4.2 mg/kg of mercury were prepared by screening to 1 inch. The coarse material was discarded, and the remaining soil was fed into a soil cleaning unit and heated to about 900°F. The cleaned soil was transferred to a soil cooling unit and prepared for reuse. The exhaust stream was sent to a dust remover and the precipitated material was sent to the soil cooling unit. The exhaust stream was then fed into an exhaust cleaning unit, which was heated to

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about 1850°F. The exhaust gas was then cooled to about 360°F by quenching with water, and subsequently an adsorption powder comprising 40% coal carbon, 10% KI_3 impregnated carbon, 40% calcium hydroxide and 10% cupric chloride impregnated carbon was added. The exhaust stream/adsorption powder mixture was fed into a baghouse, where the mercury-bound particulate was separated from the mercury-free exhaust gas. The particulate material was removed from the baghouse for analysis and disposal, while gas analyzers on the exhaust stack recorded mercury emission levels. The results of the three, two hour runs are reported in the following table.

	Run 1	Run 2	Run 3	Average
LTTD Mercury Input (lbs/hr)	0.193	0.199	0.199	0.197
Clean Soil Mercury Output (lbs/hr)	0.0020	0.0020	0.0020	0.0020
Mercury Emission Rate (lbs/hr)	0.0012	0.0006	0.0007	0.0008
Mercury Removal Efficiency[1]	99.4%	99.7%	99.6%	99.6%

[1] *Removal Efficiency (%) = (Mass Input - Stack Output- Clean Soil Output)/(Mass Input - Clean Soil Output)*

The average mercury removal efficiency of 99.6% exceeds the current mercury control efficiency specification of 96.5% efficiency, set by NJDEP air permit legislation. Mercury emissions were determined by EPA method 29-060.

Although the adsorption powder and method described and claimed are presented in the context of mercury removal from a gas stream, it should be appreciated that the powder and method are also useful for removing organics, metals and other contaminants from a gas stream.

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WHAT IS CLAIMED IS:

1. An adsorption powder for removing mercury and other metals, dioxins, furans and other organic compounds from a gas stream comprising:

a) about 1-97% carbon;

b) about 1-97% calcium hydroxide;

c) about 1-97% cupric chloride; and

d) about 1-60% KI_3 impregnated carbon.

2. The adsorption powder of claim 1 wherein the adsorption powder comprises about 10-70% carbon, about 20-80% calcium hydroxide, about 1-50% cupric chloride impregnated carbon and about 1-50% KI_3 impregnated carbon.

3. The adsorption powder of claim 1 wherein the adsorption powder comprises about 20-60% carbon, about 30-70% calcium hydroxide, about 1-30% cupric chloride impregnated carbon and about -30% KI_3 impregnated carbon.

4. The adsorption powder of claim 1 wherein the adsorption powder comprises about 25-45% carbon, about 40-60% calcium hydroxide, about 1-15% cupric chloride impregnated carbon and about 1-15% KI_3 impregnated carbon.

5. The adsorption powder of claim 1 wherein the adsorption powder comprises about 30% carbon, about 50% calcium hydroxide, about 10% cupric chloride impregnated carbon and about 10% KI_3 impregnated carbon.

6. The adsorption powder of claim 1 wherein the adsorption powder comprises about 10-70% carbon, about 20-80% calcium hydroxide, about 1-50% cupric chloride impregnated carbon.

7. The adsorption powder of claim 1 wherein the carbon is coal carbon.

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8. A method for removing mercury and other metals, dioxins, furans and other organic compounds from a gas stream using the adsorption powder of claim 1 comprising the steps of:

a) placing a solid phase mercury-containing contaminated soil feed into a rotary kiln/drum;

b) heating said kiln/drum containing said soil feed to form gaseous and solid components of the sample;

c) transferring the gaseous component of said soil feed to an exhaust cleaning unit/afterburner and the solid component of clean soil to a soil cooling unit;

d) heating the gaseous component of said contaminated soil feed in said exhaust cleaning unit/afterburner;

e) cooling the gaseous component of said contaminated soil feed;

f) adding the adsorption powder of Claim 1 to the gaseous component;

g) transferring the powder-containing gaseous component to a baghouse; and

h) releasing the substantially mercury-free gaseous component of said sample to the atmosphere.

9. The method of Claim 8 wherein the adsorption powder used in step f) comprises about 40% carbon, about 40% calcium hydroxide, about 10% cupric chloride impregnated carbon and about 10% KI_3 impregnated carbon.

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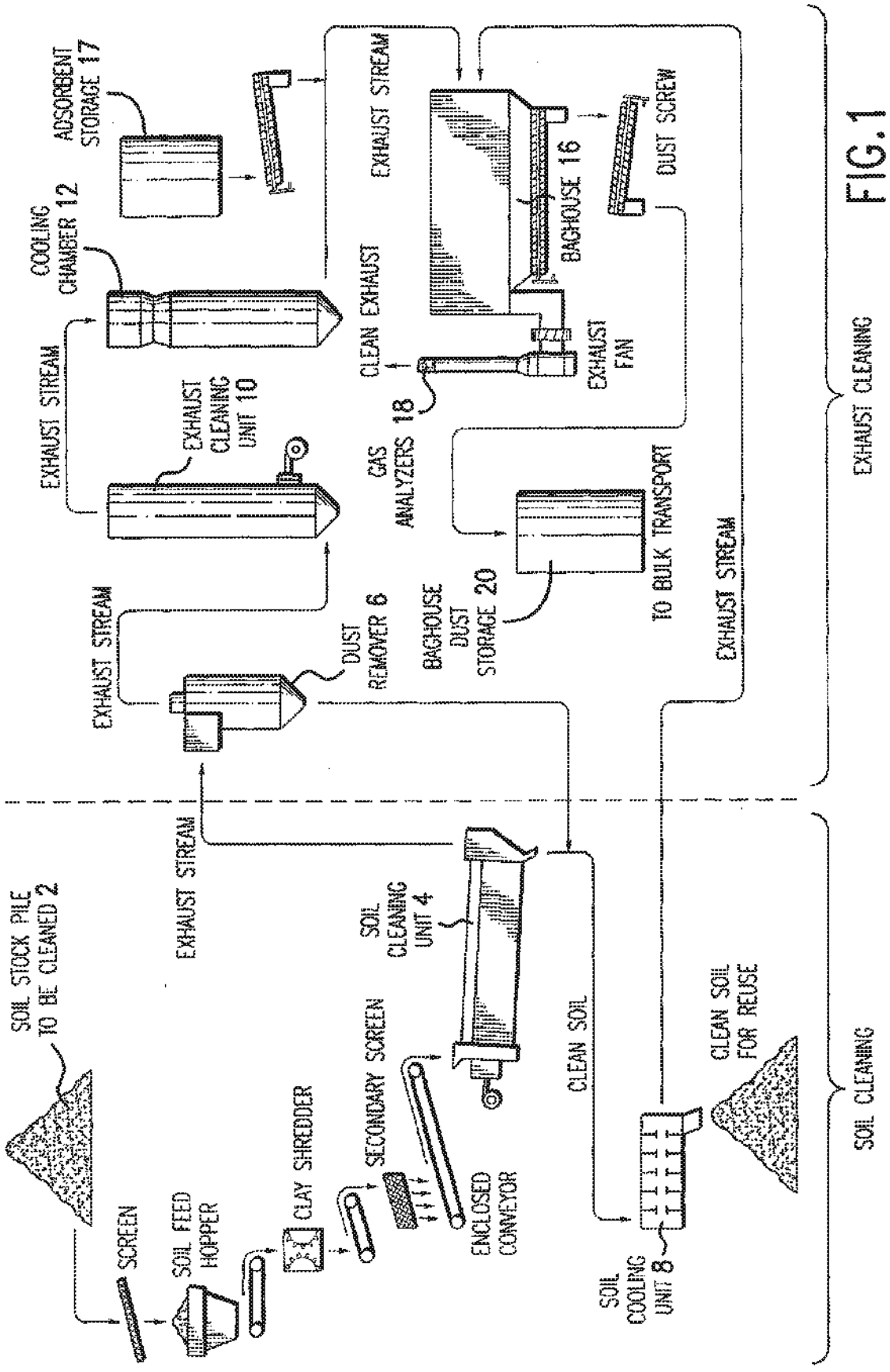

SOIL STOCK PILE TO BE CLEANED 2
SCREEN
SOIL FEED HOPPER
CLAY SHREDDER
SECONDARY SCREEN
ENCLOSED CONVEYOR
SOIL CLEANING UNIT 4
EXHAUST STREAM
DUST REMOVER 6
EXHAUST STREAM
EXHAUST CLEANING UNIT 10
EXHAUST STREAM
COOLING CHAMBER 12
ADSORBENT STORAGE 17
EXHAUST STREAM
CLEAN EXHAUST
GAS ANALYZERS 18
BAGHOUSE 16
DUST SCREW
EXHAUST FAN
BAGHOUSE DUST STORAGE 20
TO BULK TRANSPORT
EXHAUST STREAM
EXHAUST CLEANING
CLEAN SOIL
SOIL COOLING UNIT 8
CLEAN SOIL FOR REUSE
SOIL CLEANING

FIG. 1

Espacenet

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SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

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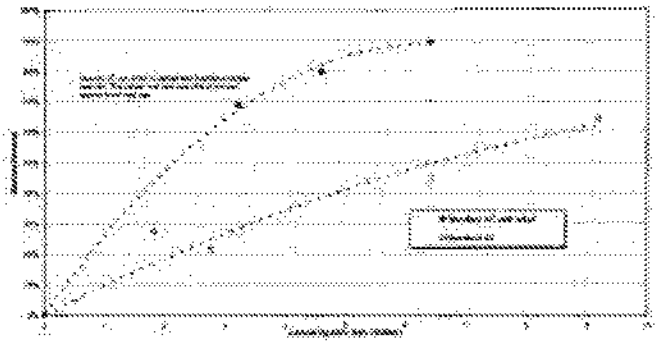
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A promoted activated carbon sorbent is described that is highly effective for the removal of mercury from flue gas streams. The sorbent comprises a new modified carbon form containing reactive forms of halogen and halides. Optional components may be added to increase reactivity and mercury capacity. These may be added directly with the sorbent, or to the flue gas to enhance sorbent performance and/or mercury capture. Mercury removal efficiencies obtained exceed conventional methods. The sorbent can be regenerated and reused. Sorbent treatment and preparation methods are also described. New methods for in-flight preparation, introduction, and control of the active sorbent into the mercury contaminated

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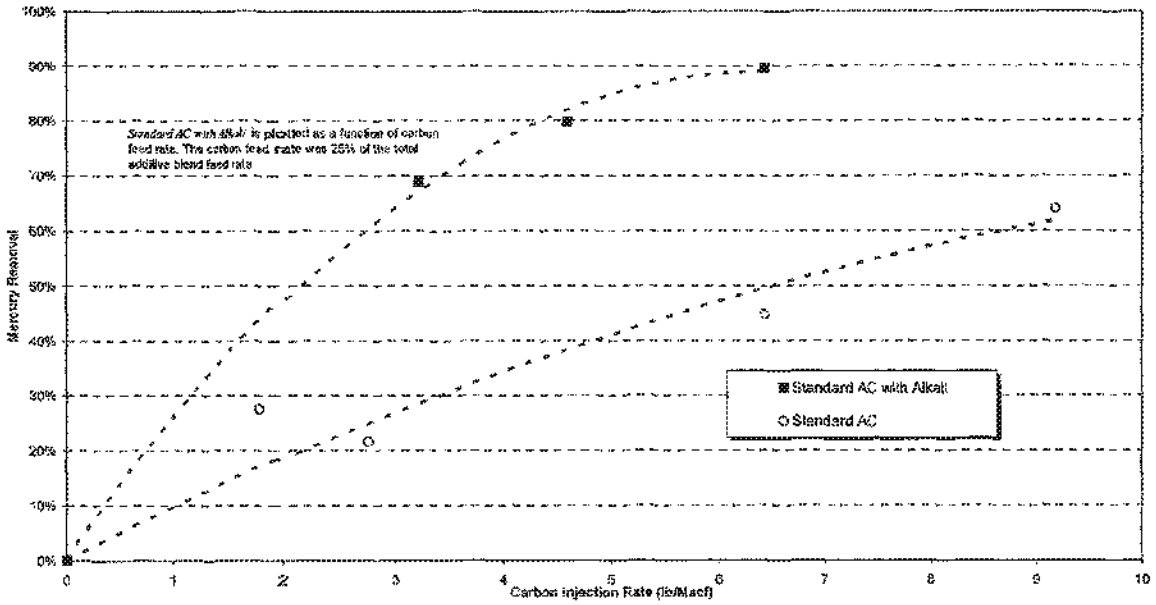
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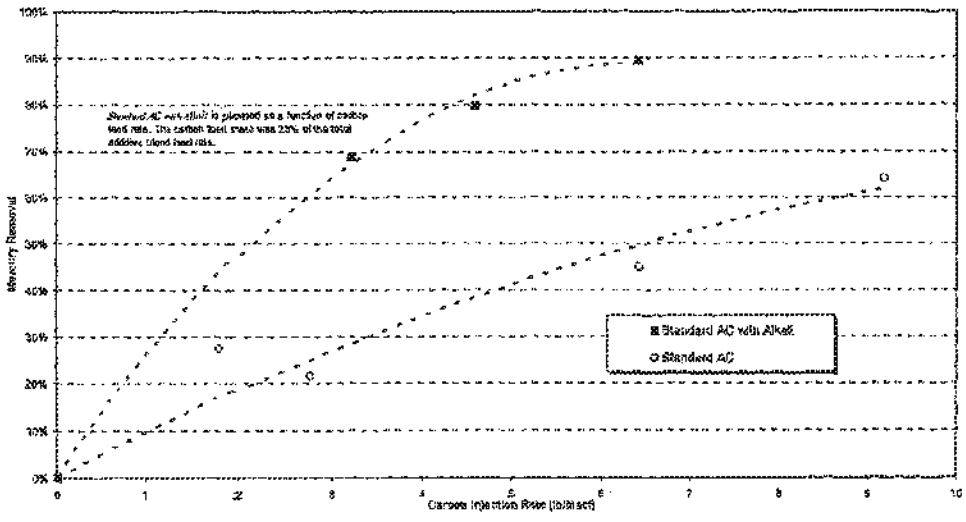
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(57) Abstract: A promoted activated carbon sorbent is described that is highly effective for the removal of mercury from flue gas streams. The sorbent comprises a new modified carbon form containing reactive forms of halogen and halides. Optional components may be added to increase reactivity and mercury capacity. These may be added directly with the sorbent, or to the flue gas to enhance sorbent performance and/or mercury capture. Mercury removal efficiencies obtained exceed conventional methods. The sorbent can be regenerated and reused. Sorbent treatment and preparation methods are also described. New methods for in-flight preparation, introduction, and control of the active sorbent into the mercury contaminated gas stream are described.

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SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

STATEMENT REGARDING FEDERALLY SPONSORED RESEARCH OR DEVELOPMENT

This invention was made with United States Government support under the U.S. Environmental Protection Agency Grant Nos. R 827649-01 and CR 830929-01. The United States Government has certain rights in this invention.

BACKGROUND OF THE INVENTION

Technical Field of the Invention

The present invention relates to methods and materials for the removal of pollutants from flue gas or product gas from a gasification system. In particular, mercury is removed from gas streams generated during the burning or gasification of fossil fuels by highly reactive regenerable sorbents.

Background of the Invention

The combustion and gasification of fossil fuel such as coal generates flue gas that contains mercury and other trace elements that originate from the fuel. The release of the mercury (and other pollutants) to the environment must be controlled by use of sorbents, scrubbers, filters, precipitators, and other removal technologies. Mercury is initially present in the elemental form during combustion and gasification. In downstream process sections, such as in the ducts and stack of a combustion system, some of the elemental mercury is oxidized. The amount that is oxidized depends on the amount of acid gases present in the flue gas and other factors. Amounts of mercury vary with the fuel, but concentrations of mercury in the stream of flue gas from coal combustion are typically less than 5 parts per billion (ppb). Large coal combustion facilities such as electric utilities may emit a pound of mercury, or more, per day. Mercury removal applications include, without limitation, flue gas from coal (or other fossil fuel) combustion, waste incineration, product gas from gasification, as well as off gases from mineral processing, metal refining, retorting, cement manufacturing, chloralkali plants, dental facilities, and crematories.

Mercury Sorbent Technologies

Several types of mercury control methods for flue gas have been investigated, including injection of fine sorbent particles into a flue gas duct and passing the flue gas through a sorbent bed. Fine-particle injection sorbents include activated carbon, metal oxide sorbent, sodium sulfide particles, and basic silicate or oxide sorbents. When particle injection is employed, the mercury captured on the sorbent particles is removed from the gas stream in a bag house or electrostatic precipitator (ESP) and collected along with ash particulate. The sulfide and basic

silicate and oxide particles are effective only for the oxidized mercury, and the metal oxide sorbents exhibit slower capture kinetics than the carbon particles. Additionally, injection of fine carbon particles into the flue gas stream has been only partially successful in removing mercury, especially elemental mercury, where effective removal of only about 60% is attained for some applications with a FF (fabric filter) to collect carbon and ash. Even lower removal rates have been observed when using an ESP to collect the carbon because the contact time of the carbon with the gas is very short.

A major problem with existing carbon injection systems is that the sorbent is initially unreactive, and only after extended exposure to the flue gas does the sorbent become effectively seasoned and provide increased reactivity with the mercury in the gas. Consequently, these sorbents must be used in large amounts, at high sorbent-to-mercury ratios, to effectively capture the mercury. These sorbents tend to be relatively expensive and cannot be easily separated from the ash for regeneration and reuse. The collection of carbon in the ash also creates solid waste disposal problems, and the spent sorbent may contaminate the collected ash, preventing its use in various applications.

Accordingly, there remains a need for more economical and effective mercury removal technology. This invention provides for cost-effective removal of pollutants including mercury, using sorbent enhancement additives and/or highly reactive sorbents, with contact times of seconds (or less), and that may be regenerated and reused.

SUMMARY OF THE INVENTION

It is thus an object of the present invention to overcome the deficiencies of the prior art and thereby to provide new and economical methods for the removal of mercury from the gases produced in the utilization of fossil fuels.

A halogen/halide promoted activated carbon sorbent is described that is highly effective for the removal of mercury from flue gas streams. The sorbent comprises a new halide-modified carbon form containing a reactive compound produced by the reaction of bromine (or halide or other halogen) with the carbon. Optional secondary components and alkali may be added to further increase reactivity and mercury capacity. Mercury removal efficiencies obtained exceed or match conventional methods with added benefits such as reduced costs. Optionally, the sorbent can be regenerated and reused. Sorbent treatment and/or preparation methods are also described. New methods for in-flight preparation, introduction, and control of the active sorbent into the mercury contaminated gas stream are described.

In some embodiments, a promoted carbon sorbent is provided comprising a base activated carbon that has reacted with a promoter selected from the group consisting of halides, halogens,

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and combinations thereof, such that the reaction product is effective for the removal of mercury from a gas stream.

In an embodiment, a promoted carbon sorbent is provided wherein the base activated carbon is selected from the group consisting of powdered activated carbon, granular activated carbon, carbon black, carbon fiber, aerogel carbon, pyrolysis char, activated carbon with an average particle size greater than that of flyash produced such that it is physically separable therefrom, and combinations thereof, and the promoter is selected from the group consisting of molecular halogens, Group V (CAS nomenclature is used throughout) halides, Group VI halides, hydrohalides, and combinations thereof. In an embodiment, the base activated carbon may have an mass mean particle diameter such that it can be substantially separated by physical means from entrained ash in the gas stream from which mercury is to be removed. In an embodiment, the base activated carbon may have a mass mean particle diameter greater than about 40 micrometers.

In another embodiment, the sorbent comprises from about 1 to about 30 grams promoter per 100 grams of base activated carbon. Another embodiment further comprises an optional secondary component comprising a halogen or a hydrohalide such that the reactivity and mercury capacity of the sorbent are enhanced.

In another embodiment, the concentration of the optional secondary component on the finished sorbent is within the range of from about 1 to about 15 wt-% of the concentration of the promoter on the finished sorbent.

In another embodiment, an optional alkali component may preferably be added to provide a synergistic effect through combination of this alkali with the primary sorbent.

In another embodiment, the optional secondary component is selected from the group consisting of Group V halides, Group VI halides, HI, HBr, HCl, and combinations thereof. In another embodiment, the promoter is substantially in vapor form when combined with the base activated carbon. In another embodiment, the promoter is combined with an organic solvent prior to reaction with the base activated carbon. In another embodiment, the promoter and optional secondary component are combined with the base activated carbon substantially simultaneously. Another embodiment further comprises adding a mercury-stabilizing reagent selected from the group consisting of S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof. Another embodiment further comprises adding an optional alkali component.

In an embodiment, a method is provided comprising providing a granular activated carbon; reacting the activated carbon with a promoter selected from the group consisting of halogens, halides, and combinations thereof, such that the reaction product comprises a promoted carbon sorbent effective for removal of mercury from a gas stream. In a further embodiment, the

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reaction product comprises from about 1 to about 30 grams promoter per 100 grams activated carbon. In another embodiment the reaction product has an average particle size distribution greater than the average size of entrained ash particles in the gas stream from which mercury is to be removed, such that the reaction product can be substantially removed from the entrained ash particles by physical means. In another embodiment the reaction product has a mass mean particle diameter greater than about 40 micrometers.

In another embodiment, the promoter is selected from the group consisting of molecular halogens, hydrohalides, Group V halides, Group VI halides, and combinations thereof. In another embodiment the promoter is in the gas phase when contacting the activated carbon. In another embodiment, the promoter is in an organic solvent when contacting the activated carbon.

In another embodiment, the promoter is selected from the group consisting of Br_2, a Group V bromide, a Group VI bromide, and combinations thereof.

In another embodiment, the method further comprises reacting the granular activated carbon with an optional secondary component comprising a halogen or a hydrohalide such that the reactivity and mercury capacity of the sorbent are enhanced. In another embodiment, the promoter and optional secondary component are contacted simultaneously with the activated carbon. In another embodiment the method further comprises adding a mercury-stabilizing reagent selected from the group consisting of S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof. In an embodiment, a method is provided for control of mercury from a flue gas at lower removal rates with substantially lower sorbent requirements. Through enhanced sorbent reactivity, mercury removal per gram of sorbent is increased, thereby decreasing the capital and operating costs by decreasing sorbent requirements.

In an embodiment, a method is provided for reducing mercury in flue gas comprising providing a sorbent, injecting the sorbent into a mercury-containing flue gas stream, collecting greater than 70 wt-% of the mercury in the flue gas on the sorbent to produce a cleaned flue gas, and substantially recovering the sorbent from the cleaned flue gas. In embodiments where less than 70 wt-% mercury removal is desired, the required removal may preferably be attained using less than half as much carbon as would be required with standard (non-enhanced) carbon. In a further embodiment, the method further comprises monitoring the mercury content of the clean flue gas, regenerating the recovered sorbent, and using the monitored mercury content of the cleaned flue gas to control the rate of injection of the sorbent. In another embodiment the injected sorbent is prepared in-flight by reacting an activated carbon and a promoter within a pneumatic transport line from which the reaction product is injected to the mercury-containing flue gas stream.

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In another embodiment, the promoter is selected from the group consisting of molecular halogens, halides, and combinations thereof. In another embodiment, the promoter is reacted in the gas phase or as a vapor. In another embodiment, the promoter is added at from about 1 to about 30 grams per 100 grams of activated carbon.

In another embodiment, the injected sorbent is prepared in-flight by reacting an activated carbon, a promoter, and an optional secondary component to enhance the reactivity and capacity of the sorbent within a pneumatic transport line from which the reaction product is injected to the mercury-containing flue gas stream.

In another embodiment, the optional secondary component is selected from the group consisting of iodine, hydrohalides, Group V halides, Group VI halides, and combinations thereof. In another embodiment, the optional secondary component is added at from about 1 to about 15 wt-% of the promoter content. In another embodiment, the method further comprises adding to the sorbent a mercury-stabilizing reagent selected from the group consisting of S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof.

In an embodiment, the method further comprises co-injecting an optional alkaline material, including without limitation alkaline and alkaline earth components, to improve the efficiency of mercury capture by capturing oxidized mercury and/or capturing gaseous components that might otherwise reduce sorbent capacity. In another embodiment, the optional alkaline material may preferably comprise calcium oxide, sodium carbonate, and the like, as are known in the art.

In another embodiment, the method further comprises using the monitored mercury content of the cleaned flue gas to control the composition of the sorbent. In another embodiment, the injected sorbent is prepared in-flight by reacting an activated carbon and a promoter within a pneumatic transport line from which the reaction product is injected to the mercury-containing flue gas stream, wherein the promoter is selected from the group consisting of molecular halogens, halides, and combinations thereof, wherein the promoter is reacted in the gas phase or as a vapor, wherein the promoter is added at from about 1 to about 30 grams per 100 grams of activated carbon, wherein the rate at which the promoter is added and the rate of sorbent injection are determined by a digital computer based at least in part on the monitored mercury content of the cleaned flue gas.

In an embodiment, a method for reducing the mercury content of a mercury and ash containing gas stream is provided wherein particulate activated carbon sorbent with a mass mean size greater than 40 μm is injected into the gas stream, mercury is removed from the gas by the sorbent particles, the sorbent particles are separated from the ash particles on the basis of size, and the sorbent particles are re-injected to the gas stream. In another embodiment, the mercury-

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containing sorbent particles are regenerated to remove some or substantially all of the mercury. In another embodiment, an alkaline component is co-injected into the gas stream. In another embodiment, the sorbent may further comprise a promoter. The promoter may preferably comprise a halide, a halogen, or both.

As will be described in more detail below, the present invention thus provides several advantages over previously known techniques, including significantly more effective and economical mercury sorbents for effluent gases, advantageously applicable to treating gas streams from fired equipment and gasification systems.

The foregoing has outlined rather broadly the features and technical advantages of the present invention in order that the detailed description of the invention that follows may be better understood. Additional features and advantages of the invention will be described hereinafter that form the subject of the claims of the invention. It should be appreciated by those skilled in the art that the conception and specific embodiments disclosed may be readily utilized as a basis for modifying or designing other structures for carrying out the same purposes of the present invention. It should also be realized by those skilled in the art that such equivalent constructions do not depart from the spirit and scope of the invention as set forth in the appended claims.

BRIEF DESCRIPTION OF THE DRAWINGS

For a more detailed description of the preferred embodiments of the present invention, reference will now be made to the accompanying drawings.

Figure 1 schematically illustrates methods for preparation of promoted carbon sorbents in accordance with the present invention.

Figure 2 illustrates a proposed mechanistic model of the chemical reactions resulting in the oxidation and capture of mercury.

Figure 3 schematically illustrates preparation of promoted carbon sorbents and processes for flue gas mercury reduction in flue gases and/or product gases from a gasification system in accordance with the present invention, including in-flight preparation of promoted carbon sorbent.

Figure 4 is a diagram illustrating breakthrough curves for 5 wt/wt% brominated NORIT Darco FGD sorbent (37 mg + 113 mg sand) in low-HCl (1 ppm) synthetic flue gas.

Figure 5 is a diagram illustrating breakthrough curves for non-halogenated NORIT Darco FGD sorbent (37 mg + 113 mg sand) in low-HCl (1 ppm) synthetic flue gas.

Figure 6 is a bar chart illustrating pilot-scale mercury removal results, including large-size sorbent results.

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Figure 7 is a diagram illustrating the effects of sorbent size and injection rate on mercury removal for ESPs and fabric filters.

Figure 8 is a diagram illustrating the breakthrough curves for a brominated NORIT Darco FGD sorbent with inert sand.

Figure 9 is a diagram illustrating the breakthrough curves for brominated NORIT Darco FGD sorbent with a co-injected alkali material.

Figure 10 is a plot of mercury removal vs. carbon injection rate with and without co-injection of alkali material.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

Herein will be described in detail specific preferred embodiments of the present invention, with the understanding that the present disclosure is to be considered an exemplification of the principles of the invention, and is not intended to limit the invention to that illustrated and described herein. The present invention is susceptible to preferred embodiments of different forms or order and should not be interpreted to be limited to the specifically expressed methods or compositions contained herein. In particular, various preferred embodiments of the present invention provide a number of different configurations and applications of the inventive method, compositions, and their uses.

The present invention provides a cost-effective way to capture pollutants by utilizing exceptionally reactive halogen/halide promoted carbon sorbents using a bromide (or other halogen/halide) treatment of the carbon, that capture mercury *via* mercury-sorbent surface reactions, at very short contact times of seconds or less. The sorbent does not require *in situ* activation (no induction period) in the gas stream to achieve high reactivity, as do conventional activated carbon sorbents. The reactivity of the sorbent toward the pollutants is greatly enhanced and the sorption capacity can be regenerated, the promoted sorbent may be regenerated, recycled and/or reused.

The treated carbons, treatment techniques, and optional additives discussed herein have applicability to mercury control from the product or effluent gas or gases from gasification systems, syngas generators, and other mercury-containing gas streams, in addition to the flue gas from combustion systems. Thus, it should be understood that the terms combustion system and flue gas as used throughout this description may apply equally to gasification systems and syngas or fuel gas, as will be understood by those skilled in the art.

Referring now to Figure 1, there is shown a block flow diagram illustrating some preferred embodiments of the process of the present invention to prepare promoted sorbents useful for mercury capture from flue gas and/or product gas form a gasification system streams. In a preferred embodiment illustrated by path 10-20, block 10 illustrates providing a base

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activated carbon, and adding a halogen or halide promoter that reacts with the carbon, illustrated at block 20, to produce a product promoted carbon sorbent. In embodiments where the halogen or halide is added, for example, as a vapor, no further steps may be necessary. In embodiments where the halogen or halide is added in, for example, a solvent, it may be desirable to employ solvent removal as illustrated by block 20A.

Referring still to Figure 1, another preferred embodiment of the process of the present invention is illustrated by path 10-20-30, comprising providing a base activated carbon as shown by block 10, adding a halogen or halide promoter that reacts with the carbon, illustrated at block 20, and adding a secondary component illustrated at block 30 that reacts with the result of block 20 to produce a product promoted carbon sorbent. In embodiments where both the halogen or halide promoter and the secondary component are added, for example, as a vapor, no further steps may be necessary. In embodiments where the halogen or halide promoter and/or secondary component are added in, for example, a solvent, it may be desirable to employ solvent removal as illustrated by block 30A.

Referring still to Figure 1, another preferred embodiment of the process of the present invention is illustrated by path 10-40, comprising providing a base activated carbon as illustrated at block 10, and adding a halogen or halide promoter and a secondary component to the activated carbon together, with which they react as illustrated by block 40, producing a product promoted carbon sorbent. As above, in embodiments where vapor additions are made to the activated carbon no further steps may be desired. In embodiments where one or more components are added in solvent, a solvent removal step may be provided as illustrated by block 40A.

Referring still to Figure 1, also illustrated are preferred embodiments in which, as illustrated by block 50, a flue gas stream is treated with product promoted carbon sorbent prepared as described above.

In some preferred embodiments the activated carbon provided may preferably be any of several types, as understood by those skilled in the art. For example, the activated carbon may include powdered activated carbon, granular activated carbon, carbon black, carbon fiber, carbon honeycomb or plate structure, aerogel carbon film, pyrolysis char, regenerated activated carbon from product promoted carbon sorbent, or other types as known in the art.

In some preferred embodiments the activated carbon provided may preferably be any of several types, as understood by those skilled in the art. For example, the activated carbon may include powdered activated carbon, granular activated carbon, carbon black, carbon fiber, carbon honeycomb or plate structure, aerogel carbon film, pyrolysis char, an activated carbon or regenerated activated carbon with a mass mean particle size greater than fly ash in a flue gas stream to be treated.

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In some preferred embodiments the activated carbon provided may preferably be any of several types, as understood by those skilled in the art. For example, the activated carbon may include powdered activated carbon, granular activated carbon, carbon black, carbon fiber, carbon honeycomb or plate structure, aerogel carbon film, pyrolysis char, an activated carbon or regenerated activated carbon with a mass mean particle diameter preferably greater than 40 micrometers, more preferably greater than 60 micrometers, or a particle size distribution greater than that of the fly ash or entrained ash in a flue gas stream to be treated, such that the activated carbon and ash can be separated by physical means.

In some preferred embodiments, the halogen or halide promoter that is added to, and reacts with, the base activated carbon may preferably comprise, by way of illustration and not limitation, a molecular halogen in vapor or gaseous form, a molecular halogen in an organic solvent, a Group V or Group VI halide, such as PBr_3 or SCl_2, respectively, in vapor, liquid, or solution form (though not in an aqueous solvent).

Embodiments are also provided in which the organic solvent may preferably comprise a chlorinated hydrocarbon, such as dichloromethane, a hydrocarbon solvent, including for example, petroleum ether, ligroin, pentane, hexane, toluene, and benzene, carbon disulfide, a waste solvent, an ether, a recycled solvent, a supercritical solvent, such as supercritical CO_2, water (though not in the case of a Group V or Group VI halide), and others as will be apparent to those of skill in the art.

Referring now to Figure 2, there is illustrated a theory developed from scientific evidence to explain the nature of the promoting compounds. For example, as illustrated in Figure 2, hydrogen bromide reacts with the unsaturated structure of the activated carbon. This may be, by way of illustration only, a carbene species on the edge of the graphene sheet structures of the carbon. Molecular bromine or a bromine compound reacts to form a similar structure, with a positive carbon that is active for oxidizing the mercury with subsequent capture by the sorbent.

It has now been found that the formation of the new bromide compound with carbon increases their reactivity toward mercury and other pollutants. Additionally, the resulting bromide compound is uniquely suited to facilitate oxidation of the mercury. The effectiveness of the oxidation apparently results from the promotion effect of the halide, exerted on the developing positive charge on the mercury during the oxidation, known in the chemical art as a specific catalytic effect. Thus, as the mercury electrons are drawn toward the positive carbon, the halide anion electrons are pushing in from the other side, stabilizing the positive charge developing on the mercury and lowering the energy requirement for the oxidation process. Bromide is especially reactive, owing to the highly polarizable electrons in the outer 4p orbitals

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of the ion. Thus, adding HBr or Br_2 to the carbon forms a similar carbon bromide, in which the positive carbon oxidizes the mercury with the assistance of the bromide ion.

Referring now to Figure 3, a schematic flow diagram is provided of mercury control system 100 comprising preparation of promoted carbon sorbents, and flue gas mercury reduction, in accordance with preferred embodiments of the present invention. There is provided base activated carbon reservoir 110, an optional halogen/halide promoter reservoir 120, an optional secondary component reservoir 130, and an optional akali component reservoir 180, each of which with corresponding flow control device(s) 201, 202, 203, and 208/209, respectively. In conjunction with the optional alkali component reservoir 180, optional flow control devices 208 and 209 can be used independently, together, or not at all.

Reservoirs 110, 120, 130, and 180 connect through their respective flow control devices and via associated piping, to transport line 115. Optional alkali component reservoir 180 may also connect, through respective flow control devices and via associated piping, to transport line 118. A source of air, nitrogen, or other transport gas(es) is provided by gas source 170 to transport line 115 for the purpose of entraining materials discharged from reservoirs 110, 120, 130, and 180 and injecting such materials, via injection point 116, into contaminated flue gas stream 15. A source of air, nitrogen, or other transport gas(es) may be provided by gas source 171 to transport line 118 for the purpose of entraining materials discharged from reservoirs 180 and injecting such materials, via injection point 119, into flue gas stream 15. Gas sources 170 and 171 may be the same or different, as desired. Alternatively, transport gas(es) may be provided to *both* transport lines 115 and 118 by gas source 170 (connection from source 170 to line 118 not shown). Although gas sources 170 and 171 are shown in Figure 3 as compressors or blowers, any source of transport energy known in the art may be acceptable, as will be appreciated by those of skill in the art.

For clarity, single injection points 116 or 119 are shown in Figure 3, although one skilled in the art will understand that multiple injection points are within the scope of the present invention. Optical density measuring device (s) 204 is connected to transport line 115 and/or 118 to provide signals representative of the optical density inside transport line 115 and/or 118 as a function of time.

Downstream from injection point 116 and 119 is provided particulate separator 140. By way of illustration and not limitation, particulate separator 140 may comprise one or more fabric filters, one or more electrostatic precipitators (hereinafter "ESP"), or other particulate removal devices as are known in the art. It should be further noted that more than one particulate separator 140 may exist, sequentially or in parallel, and that injection point 116 and 119 may be at a location upstream and/or downstream of 140 when parallel, sequential, or combinations

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thereof exist. Particulate separator 140 produces at least a predominantly gaseous ("clean") stream 142, and a stream 141 comprising separated solid materials. A sorbent/ash separator 150 separates stream 141 into a largely ash stream 152, and a largely sorbent stream 151. Stream 151 may then preferably be passed to an optional sorbent regenerator 160, which yields a regenerated sorbent stream 161 and a waste stream 162.

An optional Continuous Emission Monitor (hereinafter "CEM") 205 for mercury is provided in exhaust gas stream 35, to provide electrical signals representative of the mercury concentration in exhaust stream 35 as a function of time. The optional mercury CEM 205 and flow controllers 201, 202, 203, 208, and 209 are electrically connected via optional lines 207 (or wirelessly) to an optional digital computer (or controller) 206, which receives and processes signals and preferably controls the preparation and injection of promoted carbon sorbent into contaminated flue gas stream 15.

In operation, promoted carbon sorbent and/or an optional alkali component is injected into contaminated flue gas stream 15. After contacting the injected material with the contaminated flue gas stream 15, the injected material reduces the mercury concentration, transforming contaminated flue gas into reduced mercury flue gas, 25. The injected material is removed from the flue gas 25, by separator 140, disposed of or further separated by optional separator 150, and disposed of or regenerated by an optional regenerator 160, respectively. The reduced mercury "clean" flue gas stream 142 is then monitored for mercury content by an optional CEM 205, which provides corresponding signals to an optional computer/controller 206. Logic and optimization signals from 206 then adjust flow controllers 201, 202, 203, 208, 209 to maintain the mercury concentration in exhaust stream 35 within desired limits, according to control algorithms well known in the art. Flow controllers 201, 202, 203, 208, 209 can also be adjusted manually or be some other automated means to maintain the mercury concentration in exhaust stream 35 within desired limits, according to control algorithms well known in the art.

Referring still to Figure 3, there are illustrated several preferred embodiments for preparation and injection of promoted carbon sorbents and/or alkali components in accordance with the present invention. Stream 111 provides for introduction of base activated carbon from reservoir 110, as metered by flow controller 201 manually or under the direction of computer 206. The halogen/halide may be combined and react with the base activated carbon according to any of several provided methods. The halogen/halide may preferably be combined via line 121 directly into transport line 115, within which it contacts and reacts with the base activated carbon prior to injection point 116. This option is one form of what is referred to herein as "in-flight" preparation of a promoted carbon sorbent in accordance with the invention. Further, the halogen/halide may be combined via line 121b with base activated carbon prior to entering

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transport line 115. Still further, the halogen/halide may be contacted and react with the base activated carbon by introduction via line 121c into reservoir 110. This option is preferably employed when, for example, reservoir 110 comprises an ebullated or fluidized bed of base activated carbon, through which halogen/halide flows in gaseous form or as a vapor. Of course, the halogen/halide may also preferably be contacted with the base activated carbon in liquid form or in a solvent, as discussed previously, and solvent removal (not shown in Figure 3) may then be provided if necessary as mentioned with respect to embodiments discussed with reference to Figure 1.

Similarly, the optional secondary component may be contacted and react directly in transport line 115 via line 131, or optionally as described above with respect to the halogen/halide, via lines 131b and 131c.

Similarly, the optional alkali component from 180 may either be added in transport line 115 directly, or may be injected separately by transport line 118, combining downstream in flue gas 15 for synergistic effects with base activated carbon, promoted carbon, or optional secondary components. Being able to vary onsite the amount of the optional alkali component relative to base activated carbon, promoted carbon, or optional secondary components is a key feature to overcome and optimize for site-specific operating and flue gas conditions..

In some preferred embodiments wherein contacting between components and reaction is performed in a liquid or solvent phase, stirring of such liquid and/or slurry mixtures may be provided. In other embodiments, the halogen/halide promoter and optional secondary component(s) may preferably be sprayed in solution form into or on the base activated carbon. In some such embodiments, drying, filtering, centrifugation, settling, decantation, or other solvent removal methods as are known in the art may then be provided.

In embodiments wherein the halogen/halide promoter is in gaseous or vapor form, it may be diluted in air, nitrogen, or other gas as appropriate. The halide/halogen gas, for example, gaseous HBr or Br_2, may be passed through an ebullated or fluidized bed of granular or fibrous activated carbon, with the promoted carbon sorbent so produced removed from the top of the bed via gas entrainment for injection.

In some embodiments, the secondary component(s) may preferably comprise iodine or other halogens, hydrohalides, including without limitation HI, HBr, HCl, a Group V or Group VI element with a molecular halogen, such as SCl_2 and others. In some preferred embodiments, the promoted carbon sorbent may comprise from about 1 to about 30 g halogen/halide per 100 g base activated carbon. In some preferred embodiments, the promoted carbon sorbent may comprise an secondary component in concentration of from about 1 to about 15 wt-% of the concentration of the halogen/halide component.

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In still other embodiments, the product promoted carbon sorbent may be applied to a substrate. In other embodiments, such prepared substrate(s) may be caused to contact a contaminated flue gas or gasification system product gas stream for mercury reduction purposes. Such substrates may be monolithic, rotating, or exposed to the gas stream in any number of ways known to those skilled in the art.

In some embodiments, a method is provided whereby a mercury stabilizing reagent is added to a promoted carbon sorbent to produce a bifunctional sorbent. Such stabilizing reagent(s) may be sequentially added, either before or after the addition and reaction of the halogen/halide. In some preferred embodiments, the halogen/halide preferably comprises Br or HBr, and the mercury-stabilizing reagent may comprise S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof.

Halogens in Mercury Capture

Methodologies for using halogens for the treatment of flue gas have been problematic, owing to their reactivity with other gases and metals, resulting in corrosion and health issues. A "halogen" is defined as a member of the very active elements comprising Group VIIA (CAS nomenclature is used throughout; Group VIIA (CAS) corresponds to Group VIIB (IUPAC)) of the periodic table. In the molecular elemental form of the halogens, including F_2, Cl_2, Br_2, and I_2, the reaction with a hot flue gas components leave little to react with elemental mercury. The atomic elemental halogen form, which includes the fluorine, chlorine, bromine, and iodine atoms, is about a million times more reactive to mercury but the concentration of the atomic forms is typically extremely low. In a large portion of electric utility coal combustion facilities, the concentrations are generally not sufficient to oxidize a significant amount of mercury.

The term "halide" as used herein is defined as a compound formed from the reaction of a halogen with another element or radical. In general, halide compounds are much less reactive than the molecular halogens, having a low chemical potential. Halides are considered reduced forms that do not, alone, oxidize other compounds. In the conventional view therefore, a halide-salt-treated activated carbon will not effectively oxidize elemental mercury and capture elemental mercury.

Halogen Promoted Sorbent Characteristics

The sorbent described here has a very high initial reactivity for oxidizing mercury and therefore can be used in very small amounts to achieve very high capture efficiencies, thus lowering operation costs and lessening waste disposal problems. In addition, further disposal reductions are obtainable by regenerating and reusing the sorbents produced using the inventive technology. The time interval required for the mercury and the promoted carbon sorbents of the present invention to successfully interact in a flue gas duct, with the subsequent collection of the

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mercury on the sorbent and ash is very short – less than seconds. Clearly, such collection times require the sorbent to have both high capacity and high reactivity toward mercury. The promoted carbon sorbent can be utilized in a very finely powdered form to minimize mass transfer limitations. However, again, the reactivity should be very high to capture all of the mercury encountered by the fine particles. Additionally, use of these enhancement technologies allows capture to be effective for larger sorbent particles which also allows separation of the sorbent from the ash to enable subsequent regeneration as well as ash utilization. One feature of this invention is the process to prepare a sorbent containing a halide compound formed on the carbon structure that provides a sorbent that is highly active on initial contact with the mercury contaminated gas stream, which allows for very effective capture of the mercury.

It appears that the inventive sorbents chemically combine molecular bromine, for example, from solution, with activated carbon (edge sites). X-ray photoelectron spectroscopy has established that the addition of bromine, chlorine, HBr, or HCl formed a chemical compound in the carbon structure. Thus, the sorbent produced from halogen and activated carbon does not represent a molecular halogen form, but rather a new chemically modified carbon (or halocarbon) structure. In Addition to halide ions, this phenomenon may not occur with the less reactive iodine, where an I_2 molecular complex can exist on the carbon basal plane. In the case of bromine, the modified cationic carbon has a high chemical potential for oxidation of the mercury. Thus, an entirely new model is presented for the reactivity of the bromine-treated carbon with mercury. The reactive carbon form can preferably be generated by the addition of bromine, hydrogen bromide, or combinations of bromine and other elements, as described herein. Halogen treatment resulted in higher-activity carbons because the halide anions (especially bromide and iodide) were effective in promoting the oxidation by stabilizing the developing positive charge on the mercury in the transition state for oxidation. Based on this model, several innovative, inexpensive, activity-enhancing features have been developed.

Optional Second Component

It has been demonstrated that addition of an optional second component, in addition to the bromine, results in improved reactivity and capacity for the sorbent, typically exceeding that of both the untreated carbon and the brominated carbon. The second compound comprises either a second halogen or a compound derived from a second halogen, such as HI. Thus, in addition to having a reactive carbon form present, the second component generates a Lewis base with greater ability to stabilize the developing positive charge on the mercury. Thus, the second component is an element with more polarized electrons (4p and 5p).

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Optional Alkali Component

It has been demonstrated that addition of an optional alkali component with a base or promoted activated carbon results in improved mercury capture, typically exceeding that of both the untreated carbon and the promoted carbon. Test data indicate that flue gas contaminants, flue gas constituents (SO_2, NO_x, HCl, etc), operating temperature, mercury form, and mercury concentration may impact the effectiveness of the alkali addition. This suggests the need to be able to adjust and tailor the alkali-to-activated-carbon ratio onsite in order to overcome and optimize for a given set of site conditions.

The synergy that can be gained when co-injecting the two materials can be explained as follows. First, testing shows that binding sites on activated carbon (hereinafter "AC") can be consumed by chlorine species, sulfur species (i.e. sulfates), and other flue gas contaminants (arsenates, selenates, etc). The addition of optional alkali material will interact and react with these species/contaminants thus minimizing their consumption of AC mercury binding sites. Second, testing also shows that standard AC will continue to oxidize mercury, even though the binding sites are fully consumed. This oxidized mercury can then react with alkali material and subsequently be captured by particulate control devices. Consequently, the addition of the optional alkali component acts to protect mercury binding sites and capture oxidized mercury, thereby resulting in improved mercury reduction at lower cost. Alkali is generally much lower in cost (~ an order of magnitude less) than activated carbon, thus more of it can be used still resulting in overall lower costs.

"In-Flight" Sorbent Preparation

Furthermore, we have demonstrated that the halogen promoted carbon sorbent can be readily produced "in-flight". This is accomplished by, for example, contacting the vapors of any combination of halogens and optionally a second component, in-flight, with very fine carbon particles. The particles may be dispersed in a stream of transport air (or other gas), which also conveys the halogen/halide promoted carbon sorbent particles to the flue gas duct, or other contaminated gas stream, from which mercury is to then be removed. There is no particular temperature requirement for this contact. This technology is obviously very simple to implement, and results in a great cost savings to facilities using this technology for mercury capture.

Advantages of On-Site Preparation

In-flight preparation of the halogen/halide promoted carbon sorbent on location produces certain advantages. For example, the treatment system can be combined with the carbon injection system at the end-use site. With this technique, the halogen/halide is introduced to the carbon-air (or other gas) mixture in a transport line (or other part of the sorbent storage and injection

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system). This provides the following benefits over current conventional concepts for treating sorbents off-site:

- Capital equipment costs at a treatment facility are eliminated.
- Costs to operate the treatment facility are eliminated.
- There are no costs for transporting carbon and additive to a treatment facility.
- The inventive process uses existing hardware and operation procedures.
- The inventive technology ensures that the sorbent is always fresh, and thus, more reactive.
- No new handling concerns are introduced.
- There are no costs for removing carbon from treatment system.
- The inventive process allows rapid on-site tailoring of additive-sorbent ratios in order to match the requirements of flue gas changes, such as may be needed when changing fuels or reducing loads, thus further optimizing the economics.
- The inventive technology reduces the amount of spent sorbents that are disposed.

With the foregoing and other features in view, there is provided, in accordance with the present invention, embodiments including a process for preparing and regenerating halogen/halide promoted carbon sorbents, whose activity for mercury capture is enhanced by the addition of halogen (e.g. bromine) to the carbon structure.

Sorbent Injection Location

Some of the preferred embodiments contemplate the use of a halogen promoted sorbent in a powdered form that has been injected into a flue gas stream before or after ash particulates have been removed. Other embodiments of the inventive composition of the halogen promoted carbon sorbent comprise a powdered modified activated carbon prepared by adding Br_2 or HBr plus a second optional component. Other embodiments allow the addition of the optional alkali component in conjection with a base activated carbon and/or with the use of a halogen based sorbent and any other combinations of the sorbent technologies provided in this patent. Alternatively, embodiments include methods wherein the sorbent is on a moving contactor consisting of particles or fibers containing one or more of the compositions listed above.

Sorbent Regeneration

Any of the above embodiments of the halogen/halide promoted carbon sorbent can be easily regenerated: the poisoning contaminants from the flue gas are preferably removed and an inexpensive promoting agent added, to restore mercury sorption activity. This process of promoting the activity of the carbon itself contrasts with the earlier, more expensive, conventional methods of adding a reagent (such as peroxide, gold, triiodide, etc.) to a sorbent. The halogen/halide promoted carbon sorbent of the present invention, treated with bromine and/or

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optional components, is noncorrosive. Detailed examples of sorbent regeneration techniques are described in co-pending, commonly owned PCT patent application No. PCT/US04/12828, titled "PROCESS FOR REGENERATING A SPENT SORBENT", which is hereby incorporated by reference in its entirety.

Sorbent Injection Control Schemes

Another advantage of the present invention relates to the use of a feedback system to more efficiently utilize certain aspects of the invention. Where possible and desirable, the mercury control technology of the present invention may preferably utilize continuous measurement of mercury emissions as feedback to assist in control of the sorbent injection rate. Tighter control on the sorbent and optional component(s) levels can be achieved in this way, which will ensure mercury removal requirements are met with minimal material requirements, thus minimizing the associated costs. In an embodiment, the mercury emissions are continuously measured downstream of the injection location, preferably in the exhaust gas at the stack.

Promoted Carbon Sorbents

Reactions of halogens and acidic species with the basic binding sites on the activated carbon sorbent create sites for oxidizing mercury. Other metal ions, such as boron, tin, arsenic, gallium, Sb, Pb, Bi, Cd, Ag, Cu, Zn, or other contaminants, will also react with the oxidation sites generated on the carbon.

According to our model, adding the bromine from the bromine reagent or a proton from a hydrogen halide acid to a basic carbene site on the carbon edge structure forms a *carbocation* that accepts electrons from the neutral mercury atom forming the oxidized mercury species that is bound to the sorbent surface. The reactive site may also generate reactive bromine radicals or carbon radicals at the active sites on the carbon. Thus, the activated carbon serves to stabilize the bromine, yet provides a highly reactive bromine- containing reagent that can oxidize the mercury and promote its capture on the activated carbon. The sorbent that contains bromine is expected to be more reactive than the corresponding sorbent containing chlorine and much less expensive than the sorbent containing iodine.

EXAMPLES

To more clearly illustrate the present invention, several examples are presented below. These examples are intended to be illustrative and no limitations to the present invention should be drawn or inferred from the examples presented herein.

Example 1 -Preparation and Testing of Halogenated Carbon (& Comparative Example)

Gas Phase Halogenation

Finely powdered activated carbon (such as NORIT Darco FGD, NORIT Americas, Inc., Marshall, TX (USA), although others are suitable, as will be recognized by those skilled in the

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art), was placed in a rotating plastic barrel with side blades (a 5 ft^3 (0.14 m^3) cement mixer) fitted with a tight plastic lid to prevent loss of the fine powder during the preparation. In a separate vessel, gas phase bromine was generated by passing a nitrogen stream over a weighed amount of liquid bromine that is warmed to about 40°–50°C. The vapor pressure of the bromine was such that a dark red gas is generated a passed out of the generator. The outlet from the gaseous bromine generator is connected via a ¼ inch (0.64 cm) plastic hose to a stationary metal tube inserted through a flange in the center of the plastic lid and passing into the center of the barrel. The flange is not air tight to that the excess of nitrogen is released after the bromine is transfer to the tumbling carbon. Thus, the bromine gas stream continuously passed into the rotating barrel where it contacted the tumbling carbon. The unit is then operated until the desired amount of bromine has combined with the carbon. Typically, this is 0.4 to 1 kg of bromine to 20 kg of carbon (2–5 wt. %). When the reaction is completed, the carbon is weighed. The treated carbon is odorless and does not cause skin irritation since the bromine has completely reacted with the carbon to produce the brominated carbon.

XPS spectra demonstrate that the brominated carbon contains both covalent carbon-bound (organic) bromide as well as anionic bromide. The product contains the same moisture originally present in the activated carbon (5–17 wt%), but does not require further drying for use. The moisture is driven out at higher temperatures (>150°C), and the bromine was not released until very high temperatures

Bench-Scale Testing of Mercury Oxidation and Capture Efficiency

A bench-scale apparatus and procedure based on the above description was used to test the initial activities and capacities of several promoted activated carbon sorbents using powdered carbon, including bromine-containing activated carbons prepared from a variety of carbons, including commercially available sorbents, aerogel film sorbents, and the original precursor carbons for comparison.

A detailed description of the apparatus and its operation is provided in Dunham, G.E.; Miller, S.J. Chang, R.; Bergman, P. *Environmental Progress* **1998**, *17*. 203, which is incorporated herein by reference in its entirety. The bench scale mercury sorbent tests in the flue gas compositions were performed with finely (-400 mesh) powdered sorbents (37 mg) mixed with 113 mg sand and loaded on a quartz filter (2.5 inch (6.35 cm)). The loaded filter and holder were heated in an oven (125°C) in the simulated flue gas stream (30 SCFH (standard cubic feet/hr) or 0.79 NCMH (normal cubic meters per hour)) containing the following: O_2 (6%), CO_2 (12%), SO_2 (600 ppm), NO (120 ppm) NO_2 (6 ppm), HCl (1 ppm), Hg^0 (11 $\mu g/m^3$), H_2O (15%), and N_2 (balance). Elemental mercury was provided by a standard permeation tube source placed in a double jacketed glass condenser, and heated to the desired temperature. Mercury concentrations

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in the gas streams were determined with a continuous mercury emission monitor (Sir Galahad mercury CEM mfr. P.S. Analytical Deerfield Beach FL USA), and a $SnCl_2$ cell was used to convert oxidized species to elemental, so that both elemental and oxidized mercury concentration data could be obtained for both the influent and the effluent concentrations from the sorbent bed. Mercury concentrations were calibrated for the flow rates used. Spent sorbents were analyzed for mercury to determine the mass balance.

Referring now to Figure 4, the effluent mercury concentration data are plotted as a percent of the influent mercury versus time. The resulting curve (breakthrough curve) for the halogenated sorbents typically showed 0%–1% Hg in the effluent (99+% capture) at the beginning, and increasing only after 30–60 minutes (breakthrough point), depending on the sorbent. Figure 4 illustrates the breakthrough curves for 5 wt/wt% brominated NORIT Darco FGD sorbent (37 mg + 113 mg sand) with synthetic flue gas containing 1 ppm HCl. Total Hg (solid circles) and elemental Hg (solid squares) in the effluent are presented as a per cent of the inlet Hg. "EOT" indicates the end of test (the later data points shown are for calibration checks).

Figure 5 presents the comparative breakthrough curves for the corresponding nonhalogenated sorbents typically initiated at 5%–50% of inlet mercury, depending on the HCl concentration in the synthetic flue gas, thus indicating considerably lower reactivity for oxidation and capture of the mercury for the nonhalogenated sorbents. After breakthrough of either halogenated or nonhalogenated sorbent, most of the mercury in the effluent was oxidized mercury.

Example 2 - Gas Phase Halogenation of Fluidized Carbon

A bed of activated carbon supported in a vertical tube by a plug of glass wool was fluidized by a nitrogen stream. The top of the fluidized bed tube was connected to a catching trap for carbon fines that blow out the top of the tube. The bromine gas generator as described in Example 1 was attached to the fluidized carbon bed and the desired amount of gaseous bromine was passed into the bed. The contents of the trap were then mixed with the material in the bed and weighed. The resulting brominated carbon exhibited properties similar to the brominated carbon of Example 1.

Example 3 - Liquid Phase (water) Halogenation

A 5% solution of bromine in water was prepared by carefully adding 50 g of bromine to 1 liter of cold water. One kg of activated carbon was added to the bromine solution in a large metal can. The resulting slurry was stirred with a large paddle during the addition and for a short time afterwards until all the bromine had reacted with the carbon, as indicated by the disappearance of the red color. The slurry was then filtered using a Buchner funnel under vacuum. The moist carbon that was collected on the filter was dried in an oven at 110°C for

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several hours to constant weight. As in Example 1, some moisture remains in the carbon, however. The dried carbon was then tumbled in the rotating barrel with metal pieces to break up and fluff the carbon.

Example 4 - Addition of the Optional Second Halide Component.

Brominated carbon was produced by solution phase bromination similar to that described with reference to Example 3. However, before filtration, a solution of hydriodic acid (HI) was added to the slurry in an amount equal to 10% of the bromine amount. The slurry was stirred to complete the reaction and then filtered and dried as described in Example 3.

Example 5 - Liquid Phase Phosphohalogenation

A solution of phosphorus tribromide (500 g) in ligroin (10 liters) was stirred in a large metal can and 10 kg of activated carbon was added. The resulting slurry was stirred with a large paddle at ambient temperature to complete the reaction. The slurry was filtered under vacuum on a large Buchner funnel in several batches. The wet filter cake was dried at 110°C in an oven to constant weight. The dried product was fluffed in the rotating barrel as described in Example 3.

Example 6 - Preparation and Sorption on Larger-Particle Carbon

Tests were conducted on a pilot-scale combustor while firing a subbituminous coal, to evaluate mercury control by injecting larger-than-normal sized treated activated carbon. Standard AC sorbents generally are of fine size with a mean particle diameter of less than 20 micrometers, which is also typical of the flyash that is generated from pulverized coal combustion. Consequently, because the sizes of standard AC and flyash are similar, separation of the two is difficult. Injection of larger sized AC is generally not considered because the sorbent effectiveness decreases with size. In a scheme to recycle the injected carbon, the carbon is separated from the flyash. A separation based on size fractionation requires a treated larger particle sorbent. To test this concept, a treated larger sized (>60 μm) sorbent was developed, prepared, and tested.

Treatment - Gas Phase Halogenation

Granular activated carbon (Calgon F400) was ground and sieved through conventional mesh screens. The mesh size fraction -170 to +240 (corresponding to about 60 to about 88 micrometers) was collected and placed in a rotating vessel as described in Example 1 above. In a separate vessel, gas phase bromine was generated by passing a nitrogen stream over a weighed amount of liquid bromine that was warmed to about 40°–50°C, and the outlet from this gaseous bromine generator was connected via a 1/4 inch (6.35 mm) plastic hose to a stationary metal tube inserted through a flange in the center of the lid and passing into the center of the rotating vessel, also as described in Example 1. The unit was operated until the desired amount of bromine had

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combined with the carbon, in this case 0.05 kg of bromine to 1 kg of carbon (5 wt. %). When the reaction was completed, the carbon was weighed. The treated carbon was odorless as has been described above.

PTC Apparatus

The pilot-scale combustor, known as the "Particulate Test Combustor" (hereinafter "PTC"), is a 550,000-Btu/hr (about 161 kW) pulverized coal ("PC")-fired unit, designed to generate combustion flue gas properties and fly ash that are representative of those produced in a full-scale utility boiler. The combustor is oriented vertically to minimize wall deposits. A refractory lining helps to ensure adequate flame temperature for complete combustion and prevents rapid quenching of the coalescing or condensing fly ash. Based on the superficial gas velocity, the mean residence time of a particle in the combustor is approximately 3 seconds. The coal nozzle of the PTC fires axially upward from the bottom of the combustor, and secondary air is introduced concentrically to the primary air with turbulent mixing. Coal is introduced to the primary air stream via a screw feeder and eductor. An electric air preheater is used for precise control of the combustion air temperature. Originally, the PTC used cold-water annular heat exchangers to provide flue gas temperature control to the baghouse (also referred to as a "fabric filter") or electrostatic precipitator (ESP). However, analysis of ash deposits collected from the heat exchangers indicated that some mercury was collected on the duct walls. To minimize this effect, the heat exchangers were modified to provide for higher duct wall temperatures.

The PTC instrumentation permits system temperatures, pressures, flow rates, flue gas constituent concentrations, and particulate control device (baghouse, Advanced Hybrid Particle Collector/AHPC™, and/or electrostatic precipitator/ESP) operating data to be monitored continuously and recorded on a data logger.

PTC Procedure

Flue gas samples were taken at combinations of two of the three available system sample points: the furnace exit, the particulate control device inlet, and the particulate control device outlet. After passing through sample conditioners to remove moisture, the flue gas was typically analyzed for O_2, CO, CO_2, SO_2, and NO_x. Each constituent was normally analyzed at both the furnace exit and the outlet of the particulate control device simultaneously, using two analyzers. The concentration values from all of the instruments were recorded continuously. In addition, data were manually recorded at set time intervals. NO_x was determined using a pair of Rosemount Analytical NO_x chemiluminescent analyzers. SO_2 was measured using a pair of Ametek Instruments photometric gas analyzers. The remaining gases were measured by a pair of Rosemount Analytical multi-gas continuous emissions monitors. Each of these analyzers was regularly calibrated and maintained to provide accurate flue gas concentration measurements.

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The baghouse vessel was a 20 inch (50.8 cm) (ID) chamber that is heat-traced and insulated, with the flue gas introduced near the bottom. The combustor produced about 200 ACFM (actual cubic feet per minute; about 5.7 actual m^3/min) of flue gas at 300°F (about 150°C), therefore three 13-ft by 5-inch (3.96 m by 12.7 cm) bags provided an air-to-cloth ratio of 4 ft/min (1.22 m/min). Each bag was cleaned separately in operation with its own diaphragm pulse valve. In order to quantify differences in pressure drop for different test conditions, the bags were cleaned on a time basis, rather than with the cleaning cycle initiated by pressure drop. Once bag cleaning was initiated, all three bags were pulsed in rapid succession on-line.

Tests were also conducted with a single-wire, tubular ESP replacing the fabric filter. The ESP unit was designed to provide a specific collection area of 125 at 300°F (150°C). Since the flue gas flow rate for the PTC is 130 SCFM (standard cubic feet per minute; about 3.7 NCMM (normal m^3/min)), the gas velocity through the ESP is 5 ft/min (about 1.52 m/min). The plate spacing for the ESP unit is 11 in (27.9 cm). The ESP was designed to facilitate thorough cleaning between tests so that all tests can begin on the same basis.

PTC Results

Results are illustrated in Figure 6. As can be observed in Figure 6, even though the tested sorbent particle size is significantly larger than normal sorbent particles, the treated larger-than-normal sized (that is, >60 micrometers) activated carbon sorbent was quite effective at capturing mercury. Approximately 75% of the mercury was captured when the larger-sized treated AC was injected ahead of the pilot-scale ESP, while approximately 85% of the mercury was captured when injected ahead of the pilot-scale fabric filter ("FF"). Note that in Figure 6 (and throughout) "Macf" (and "MACF") indicates million actual cubic feet (1 MACF is about 0.028 million actual cubic meters or "MACM").

Referring now to Figure 7, it can be observed that the larger-sized treated AC when injected ahead of the pilot-scale ESP (diamond symbol(s)) performed better than the finer standard AC (triangles) under the same arrangement. In comparison, when injected ahead of the fabric filter (FF), the larger-sized treated AC (square) performed similarly to slightly worse. However, for this application, the larger-sized treated AC can be physically separated from the smaller flyash particles, and the sorbent can then be regenerated, recycled, and reused. This will substantially improve overall utilization and economics. These data thus show that a larger-than-normal sized sorbent can provide effective mercury control and ease flyash and AC separation, thereby also preserving the characteristics of the flyash for sale and beneficial use. Accordingly, because >60 μm sorbent particles have been successfully demonstrated, superior mercury control can be obtained with >40 μm particles, which may be preferred in some applications, depending

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on the sorbent particle/ash separation system used. Note that in Figure 7 (and throughout) "Macf" (and "MACF") indicates million actual cubic feet.

Example 7 - Liquid Phase (Organic Solvent) Halogenation

A 5% solution of bromine in ligroin was prepared by carefully adding 50 g of bromine to 1 liter of cold ligroin. One kg of activated carbon was added to the bromine solution in a large metal can. The slurry was stirred with a large paddle during the addition and for a short time afterwards until all the bromine had reacted with the carbon as indicated by the disappearance of the red color. The slurry was filtered using a Buchner funnel under vacuum. The carbon cake that was collected on the filter was dried in an oven at 110°C for several hours until it appeared dry and a constant weight was obtained. As in Example 1, some moisture was left in the carbon, however. The dried carbon was then tumbled in the rotating barrel with metal pieces to break up and fluff the carbon.

Example 8 - Promoted Activated Carbon Sorbents

A bench-scale procedure based on the above description was used to test the initial activities and capacities of several promoted activated carbon sorbents using powdered carbon, including the bromine-containing activated carbons prepared from a commercially available sorbent and an aerogel carbon film sorbent, as well as the original precursor carbons for comparison. Bromine-treated carbons were prepared by impregnation of the powdered activated carbon precursors in a stirred solution of bromine in carbon tetrachloride or methylene chloride, or alternatively, in an aqueous solution of HBr, followed by drying in air at ambient temperature and drying in an oven at 100°C in air or nitrogen. Bromine-treated carbons were also prepared by impregnating bromine from the gas phase by passing the gas through a rotating dry bed of the activated carbon precursor. The results indicated that adding a second component to the solution improved the capacity of the sorbent.

The carbons were initially tested in a heated bed, where a synthetic flue gas stream containing elemental mercury (11 $\mu g/m^3$) was passed through the bed. Concentrations of total and elemental Hg in the effluent gas were determined using a Sir Galahad mercury CEM ("continuous emission monitor") (mfr. P S Analytical, Deerfield Beach, FL, USA). The powdered sorbent was supported on a quartz filter during the test, and the other sorbents were tested as a triple layer. A comparison of the original commercial-grade powdered carbon sorbent with the sorbent after it was treated with 0.1 N HBr, and the powder was collected by centrifugation and drying, revealed that the mercury capture activity increased from an initial capture efficiency of about 50% of the Hg in the inlet to 100% capture. A comparison of the sorbent after subsequent regeneration with HBr indicated that it not only captured mercury at the same level as before (100% capture) but its capacity was prolonged by several minutes, and thus

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enhanced. Similar results were obtained with the carbon film and carbon fiber sorbents by treatment with molecular bromine in solution or in dry beds as described above.

Example 9 - Fluidized/Ebullated Bed Preparation

An activated carbon sorbent was prepared by treating the carbon by impregnating molecular bromine from a gas composition containing molecular bromine by flowing the gas through a liquid bromine reservoir in series with a fluidized bed or ebullated bed of the carbon. The amount of bromine taken up by the carbon ranges (in one example) from <1 to about 30 g per 100 g of activated carbon, depending on the proportions used.

Example 10 - Full-Scale Testing

In this example, a baghouse (fabric filter) or ESP was used to collect particulates in the exhaust of a full-scale commercial pulverized coal-burning facility. A scrubber and sorbent bed were also used to remove undesired constituents from the flue gas stream, before being fed to the stack. In this example, the halogen/halide promoted carbon sorbent was injected into the flue gas after the boiler. In general however, the inventive sorbent can be injected where desired (e.g., before, after, or within the boiler).

In one exemplary test conducted at a facility fired with lignite coal, the flue gas phase mercury (elemental) concentration was between 10 and 11 $\mu g/m^3$. The ash and injected carbon were collected in the baghouse at 350°F to 375°F (about 175-190°C). Injection of commercial-grade activated carbon powder (untreated) at a rate of 1.0 lb/MACF ("MACF" and "Macf" represent one million actual cubic feet; 1.0 lb/MACF is about 16 kg/MACM (million actual cubic meters)) resulted in mercury effluent concentrations of 3.8-4.2 $\mu g/m^3$ (representing 62%-58% removal of the mercury from the gas, respectively), and at 2.0 lb/MACF (about 32 kg/MACM), gave 74%-71% removal. Injection of the bromine-treated carbon at 1.0 lb/MACF resulted in 73%-69% removal and at 2.0 lb/MACF gave 86%-84% removal. Thus, a significant increase in the mercury capture was exhibited during use of the bromine promoted carbon sorbent of the present invention.

Example 11A - Addition of Optional Alkaline Component – Bench-Scale

The efficiency of the activated carbons for mercury capture can be improved considerably by employing a basic material co-injected with the activated carbon, in order to capture any oxidized mercury that may be released from the sorbent, or to capture some of the sulfur or selenium oxides in the flue gas that can have a detrimental effect on the sorbent capacity.

Bench-scale testing was conducted by preparing a filter composed of 37 mg of brominated activated carbon mixed with 113 mg of calcium oxide. The test was conducted as described in Example 1 and compared with the same carbon sorbent but with an inert diluent. The breakthrough curve for the mixture of brominated (2%) NORIT Darco FGD sorbent with

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inert sand is shown in Figure 8, and the breakthrough curve for the mixture with CaO is shown in Figure 9. It can be seen that the point of 50% breakthrough improves to 65 minutes with the mixture with CaO from only 48 min with the sand mixture.

Example 11B - Addition of Optional Alkaline Component – Pilot-Scale

Tests were conducted on the pilot-scale PTC combustor described above with reference to Example 6 while firing a Texas lignite to evaluate mercury control by co-injecting a standard activated carbon (also referred to herein as "AC") and an alkali material upstream of a fabric filter. Typical results are illustrated in Figure 10. As shown in Figure 10, co-injecting lime with activated carbon vastly improved mercury removal. Mercury removals of approximately 90% were achieved with the co-injected sorbents, whereas less than 60% removal was achieved with the use of standard AC alone, even at much higher injection rates. Data from similar tests show that injecting similar quantities of sodium carbonate and AC, and lime and AC, resulted in mercury removals of approximately 80%, and 87%, respectively. These data suggest that other alkali can also be co-injected with AC to improve mercury removal. Other data show that flue gas temperature may impact the effectiveness of the alkali addition. Further test data indicate that flue gas contaminants, flue gas constituents (SO_2, NO_x, HCl, etc.), operating temperature, mercury form, and mercury concentration may impact the effectiveness of the alkali addition. This indicates that it may be desirable to be able to adjust and tailor, onsite, the alkali-to-AC ratio in order to optimize removal for a given set of site conditions.

Without wishing to be bound by any particular theory, the synergy observed in the improved performance when co-injecting the two materials can be explained as follows. First, tests indicate that binding sites on AC can be consumed by sulfur species and other contaminants. The alkali material interacts and reacts with these species thus minimizing their consumption of AC mercury binding sites. Second, other work has shown that standard AC will continue to oxidize mercury even though the binding sites are fully consumed. This oxidized mercury can then react with alkali material and subsequently be captured by the particulate control device. Thus, combining alkali with treated and/or non-treated AC synergistically takes advantage of these two mechanisms, resulting in improved mercury capture at reduced costs.

Example 12- Brominated Carbon Sorbent for Gasification Fuel Gas

Preparation of 5%Br2W-AC

Using a procedure similar to Example 3, a 2.5 wt/vol% solution of bromine in water was prepared. Granular Calgon F400 was added to the bromine solution to give a 5 wt/wt% brominated carbon product. The bromine solution was stirred with a large paddle during and after the addition until the red color in the water disappeared. The suspension was filtered by

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vacuum on a large Buchner funnel. The filter cake was dried in air, and then in an oven at 110°C until a stable weight was obtained The moisture was reduced to 15%.

Preparation of 5%Br2D-AC

A brominated sorbent was prepared from Br_2 addition in solvent as described in Example 7, except that dichloromethane was used as the solvent instead of ligroin, and granular Calgon F400 was used.

Preparation of 5%PBr3-AC

A phosphohalogenated sorbent was prepared from PBr_3 using the method described in Example 5, except granular Calgon F400 was used.

Testing in Hydrogen Atmosphere - Procedure

To simulate the capture of mercury from a heated fuel gas or syngas from coal gasification, tests were conducted employing a stream comprising 10% vol/vol hydrogen in nitrogen passing through the sorbent at 500 cc/min. The stream contained 26.9 micrograms/m^3 of elemental mercury from a commercial mercury permeation source.

In the tests, the sorbent (0.5 g) was placed in a 0.39 inch (1 cm, inside diameter) glass tube fitted with a medium frit sintered glass filter disc to hold the sorbent in the gas stream. The tube containing the sorbent bed was connected to a gas inlet tube for introducing the gas stream containing the mercury vapor and at the outlet to a tube connection to the detector. The detector was a Semtech 2000 continuous mercury emission monitor. The tube was equilibrated in a nitrogen flow (450 cc/min) for 5 minutes at ambient temperature to stabilize the system. The detector showed 0 concentration of mercury in the effluent from the sorbent bed. (The blank run with no sorbent read 26.9 micrograms/m^3). The tube was then placed in an oven at the selected temperature for the test (from 250° to 400°C). Effluent mercury concentration data from the detector were collected until the detector showed a constant reading for 5 minutes. Hydrogen (50 cc/min) was then added to the gas stream and detector readings were taken every 5 min. Tests were conducted at several oven temperatures for various periods of time up to 3 hours, depending on the temperature and sorbent. The elemental mercury concentration data were plotted as a percent of inlet mercury concentration versus time as in Example 1. All the mercury in the effluent was elemental, so a single detector was sufficient, and no $SnCl_2$ trap was needed to convert to elemental mercury (as in Example 1). The time for 50% breakthrough (time to reach 50% capture) was then determined from the breakthrough curves.

Results

The results are shown in Table 1 (below) for the unbrominated sorbent (Calgon F-400), the brominated sorbents (5%Br2W-AC and 5%BrD-AC), and the phosphobrominated sorbent

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(5%PBr3-AC). The maximum mercury concentration obtained in the effluent in each run is also reported in Table 1 for the time period indicated in the last column.

Under the reducing hydrogen conditions, the unbrominated sorbent broke through immediately and was exhausted after only 6.5 min. This complete failure occurred because the hydrogen reduces the captured mercury in the unbrominated sorbent at any temperature above 100°C. Both of the brominated sorbents exhibited excellent reactivity and good capacity at all temperatures, up to at least 400°C. The phosphobrominated sorbent exhibited superior reactivity and capacity at all temperatures, up to at least 400°C.

Table 1

Times for 50% Breakthrough
Maximum Observed Hg Concentrations for Sorbents
(10% Hydrogen Streams)

Sorbent	Temp (°C)	50% breakthrough (min)	Maximum [Hg] ($\mu g/m^3$)	Time (min)
F-400	250	6	20.3	6.5
5%Br2W-AC	250	>150	1.4	150
5%Br2W -AC	300	>180	4.3	180
5%Br2W-AC	350	160	15.1	180
5%Br2W-AC	400	60	13.9	65
5%PBr3-AC	250	>140	0.4	140
5%PBr3-AC	300	>150	0.5	150
5%PBr3-AC	350	>150	1.4	150
5%Br2D-AC	350	>180	2.1	180
5%Br2D-AC	400	>180	10.9	180

While the preferred embodiments of the invention have been shown and described, modifications thereof can be made by one skilled in the art without departing from the spirit and teachings of the invention. The embodiments described herein are exemplary only, and are not intended to be limiting. Many variations and modifications of the invention disclosed herein are possible and are within the scope of the invention. Accordingly, the scope of protection is not limited by the description set out above, but is only limited by the claims which follow, that scope including all equivalents of the subject matter of the claims.

The examples provided in the disclosure are presented for illustration and explanation purposes only and are not intended to limit the claims or embodiment of this invention. While the preferred embodiments of the invention have been shown and described, modifications

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thereof can be made by one skilled in the art without departing from the spirit and teachings of the invention. Process criteria, equipment, and the like for any given implementation of the invention will be readily ascertainable to one of skill in the art based upon the disclosure herein. The embodiments described herein are exemplary only, and are not intended to be limiting. Many variations and modifications of the invention disclosed herein are possible and are within the scope of the invention. Use of the term "optionally" with respect to any element of the invention is intended to mean that the subject element is required, or alternatively, is not required. Both alternatives are intended to be within the scope of the invention.

The discussion of a reference in the Background is not an admission that it is prior art to the present invention, especially any reference that may have a publication date after the priority date of this application. The disclosures of all patents, patent applications, and publications cited herein are hereby incorporated herein by reference in their entirety, to the extent that they provide exemplary, procedural, or other details supplementary to those set forth herein.

Although the invention is described herein as a sorbent material and associated processes for its preparation and use, it is nevertheless not intended to be limited to the details described, 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.

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WE CLAIM:

1. A promoted carbon sorbent comprising a base activated carbon that has reacted with a promoter selected from the group consisting of halides, halogens, and combinations thereof, such that the reaction product is effective for the removal of mercury from a gas stream.

2. The promoted carbon sorbent of claim 1 wherein the base activated carbon is selected from the group consisting of powdered activated carbon, granular activated carbon, carbon black, carbon fiber, aerogel carbon, pyrolysis char, activated carbon with an average particle size greater than 40 micrometers, and combinations thereof, and the promoter is selected from the group consisting of molecular halogens, Group V halides, Group VI halides, hydrohalides, and combinations thereof.

3. The promoted carbon sorbent of claim 2 wherein the sorbent comprises from about 1 to about 30 grams promoter per 100 grams of base activated carbon.

4. The promoted carbon sorbent of claim 3 further comprising an optional secondary component comprising a halogen or a hydrohalide such that the reactivity and mercury capacity of the sorbent are enhanced.

5. The promoted carbon sorbent of claim 4 further comprising an optional alkaline component selected from the group consisting of alkali elements, alkaline earth elements, alkali salts, alkaline earth salts, and combinations thereof.

6. The promoted carbon sorbent of claim 4 wherein the concentration of the optional secondary component on the finished sorbent is within the range of from about 1 to about 15 wt-% of the concentration of the promoter on the finished sorbent.

7. The promoted carbon sorbent of claim 6 wherein the optional secondary component is selected from the group consisting of Group V halides, Group VI halides, HI, HBr, HCl, and combinations thereof.

8. The promoted carbon sorbent of claim 7 wherein the optional secondary component is selected from the group consisting of I_2, HI, and combinations thereof.

9. The promoted carbon sorbent of claim 8 wherein the promoter is substantially in vapor form when combined with the base activated carbon.

10. The promoted carbon sorbent of claim 8 wherein the promoter is combined with an

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organic solvent prior to reaction with the base activated carbon.

11. The promoted carbon sorbent of claim 8 wherein the promoter and optional secondary component are combined with the base activated carbon substantially simultaneously.

12. The promoted carbon sorbent of claim 8 further comprising a mercury-stabilizing reagent selected from the group consisting of S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof.

13. The promoted carbon sorbent of claim 12 wherein the promoter is selected from the group consisting of Br_2, HBr, and combinations thereof.

14. The promoted carbon sorbent of claim 3 further comprising a mercury-stabilizing reagent selected from the group consisting of S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof.

15. The promoted carbon sorbent of claim 14 further comprising an optional alkaline component comprising an alkaline or alkaline earth element wherein the promoter is selected from the group consisting of Br_2, HBr, and combinations thereof.

16. A method comprising providing a granular activated carbon; reacting the activated carbon with a promoter selected from the group consisting of halogens, halides, and combinations thereof, such that the reaction product comprises a promoted carbon sorbent effective for removal of mercury from a gas stream.

17. The method of claim 16 wherein the reaction product comprises from about 1 to about 30 grams promoter per 100 grams activated carbon.

18. The method of claim 16 wherein the reaction product has an average particle size distribution greater than about 40 micrometers.

19. The method of claim 16 wherein the promoter is selected from the group consisting of molecular halogens, hydrohalides, Group V halides, Group VI halides, and combinations thereof.

20. The method of claim 19 wherein the promoter is in the gas phase when contacting the activated carbon.

21. The method of claim 19 wherein the promoter is in an organic solvent when contacting the activated carbon.

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22. The method of claim 20 wherein the promoter is selected from the group consisting of Br_2, a Group V bromide, a Group VI bromide, and combinations thereof.

23. The method of claim 18 further comprising providing an optional alkaline component to improve the effectiveness of mercury removal.

24. The method of claim 20 further comprising reacting the granular activated carbon with an optional secondary component comprising a halogen or a hydrohalide such that the reactivity and mercury capacity of the sorbent are enhanced.

25. The method of claim 24 further comprising providing an optional alkaline component to improve the effectiveness of mercury removal.

26. The method of claim 24 wherein the promoter and optional secondary component are contacted simultaneously with the activated carbon.

27. The method of claim 24 wherein the optional secondary component comprises I_2 or HI.

28. The method of claim 20 further comprising adding a mercury-stabilizing reagent selected from the group consisting of S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof.

29. The method of claim 24 further comprising adding a mercury-stabilizing reagent selected from the group consisting of S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof.

30. A method for reducing mercury in flue gas comprising providing a sorbent, injecting the sorbent into a mercury-containing flue gas stream, collecting greater than 70 wt-% of the mercury in the flue gas on the sorbent to produce a cleaned flue gas, and substantially recovering the sorbent from the cleaned flue gas.

31. The method of claim 30 further comprising co-injecting an optional alkaline component into the mercury containing flue gas stream.

32. The method of claim 30 further comprising monitoring the mercury content of the clean flue gas, regenerating the recovered sorbent, and using the monitored mercury content of the cleaned flue gas to control the rate of injection of the sorbent.

33. The method of claim 30 wherein the injected sorbent is prepared in-flight by reacting an activated carbon and a promoter within a pneumatic transport line from which the reaction product is injected to the mercury-containing flue gas stream.

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34. The method of claim 33 wherein the promoter is selected from the group consisting of molecular halogens, halides, and combinations thereof.

35. The method of claim 34 wherein the promoter is selected from the group consisting of Br_2, HBr, Group V bromides, Group VI bromides, and combinations thereof.

36. The method of claim 35 wherein the promoter is reacted in the gas phase or as a vapor.

37. The method of claim 36 wherein the promoter is added at from about 1 to about 30 grams per 100 grams of activated carbon.

38. The method of claim 37 wherein the injected sorbent is prepared in-flight by reacting an activated carbon, a promoter, and an optional secondary component to enhance the reactivity and capacity of the sorbent within a pneumatic transport line from which the reaction product is injected to the mercury-containing flue gas stream.

39. The method of claim 38 wherein the optional secondary component is selected from the group consisting of iodine, hydrohalides, Group V halides, Group VI halides, and combinations thereof.

40. The method of claim 39 further comprising co-injecting an optional alkaline component into the mercury containing flue gas stream.

41. The method of claim 39 wherein the optional secondary component is added at from about 1 to about 15 wt-% of the promoter content.

42. The method of claim 41 wherein the optional secondary component is selected from the group consisting of I_2, HI, and combinations thereof.

43. The method of claim 42 wherein the optional secondary component is added in the gas phase or as a vapor.

44. The method of claim 41 further comprising adding to the sorbent a mercury-stabilizing reagent selected from the group consisting of S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof.

45. The method of claim 32 further comprising using the monitored mercury content of the cleaned flue gas to control the composition of the sorbent.

46. The method of claim 40 further comprising monitoring the mercury content of the clean

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flue gas and using the monitored mercury content to control the composition of the sorbent and the alkaline-to-sorbent injection ratio.

47. The method of claim 45 wherein the injected sorbent is prepared in-flight by reacting an activated carbon and a promoter within a pneumatic transport line from which the reaction product is injected to the mercury-containing flue gas stream, wherein the promoter is selected from the group consisting of molecular halogens, halides, and combinations thereof, wherein the promoter is reacted in the gas phase or as a vapor, wherein the promoter is added at from about 1 to about 30 grams per 100 grams of activated carbon, wherein the rate at which the promoter is added and the rate of sorbent injection are determined by a digital computer based at least in part on the monitored mercury content of the cleaned flue gas.

48. A method for reducing the mercury content of a gas stream comprising the steps of injecting a powdered activated carbon sorbent having a mass-mean particle size >40μm into a gas stream containing mercury and ash particles, capturing mercury from the gas stream on the sorbent particles, mechanically separating the sorbent particles from the ash particles, and re-injecting the separated sorbent particles into the gas stream.

49. The method of claim 48 further comprising regenerating the sorbent particles to substantially remove contaminants therefrom to recover capacity for capturing mercury.

50. The method of claim 48 further comprising co-injecting into the gas stream an alkaline component selected from the group consisting of alkali elements, alkaline earth elements, alkali salts, alkaline earth salts, and combinations thereof.

51. The method of claim 48 wherein the powdered activated carbon sorbent further comprises a promoter selected from the group consisting of halides, halogens, and combinations thereof.

52. The method of claim 51 further comprising co-injecting into the gas stream an alkaline component selected from the group consisting of alkali elements, alkaline earth elements, alkali salts, alkaline earth salts, and combinations thereof.

53. The method of claim 48 wherein the particulate activated carbon sorbent has a mass-mean particle size >60μm.

54. The method of claim 52 wherein the particulate activated carbon sorbent has a mass-mean particle size >60μm.

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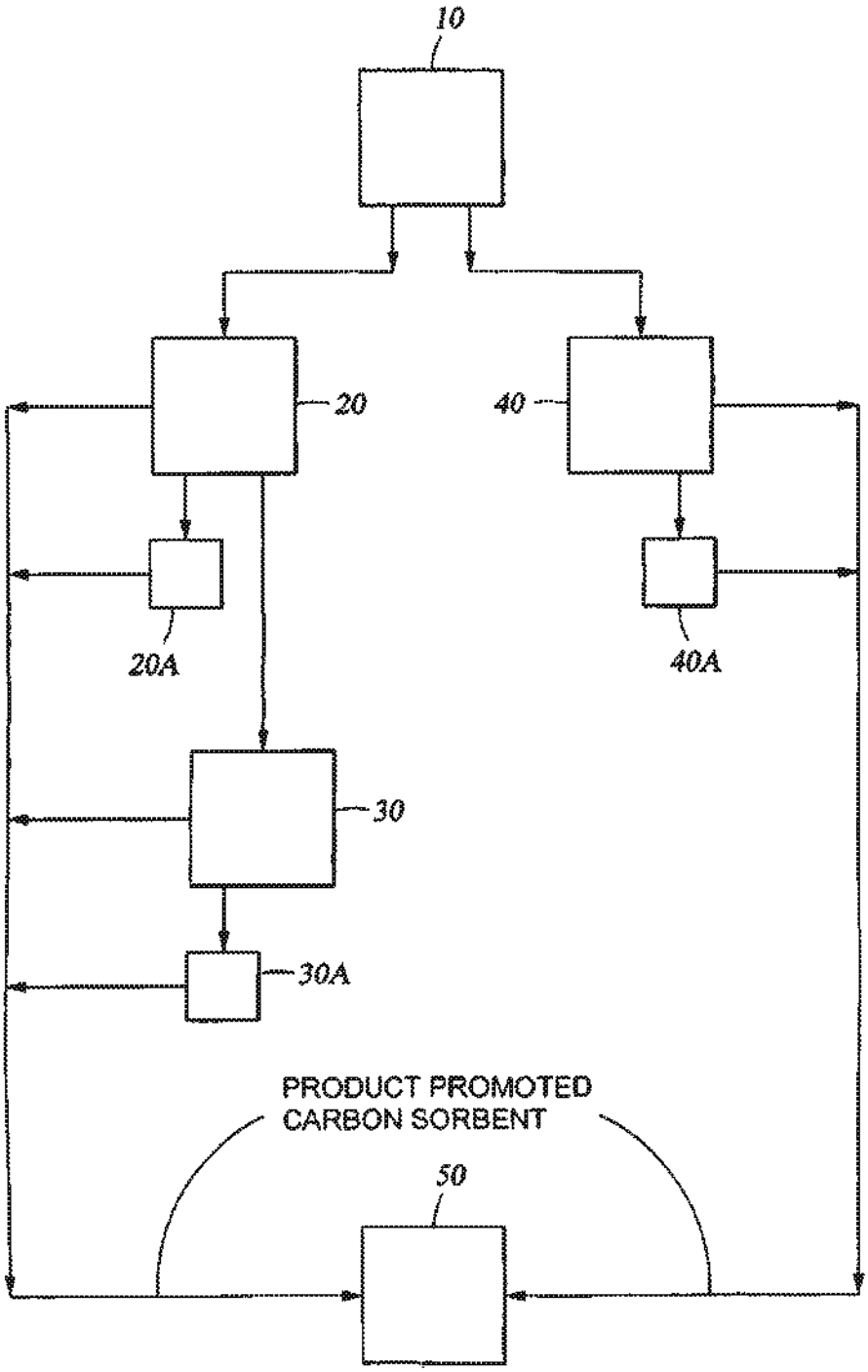


Fig. 1

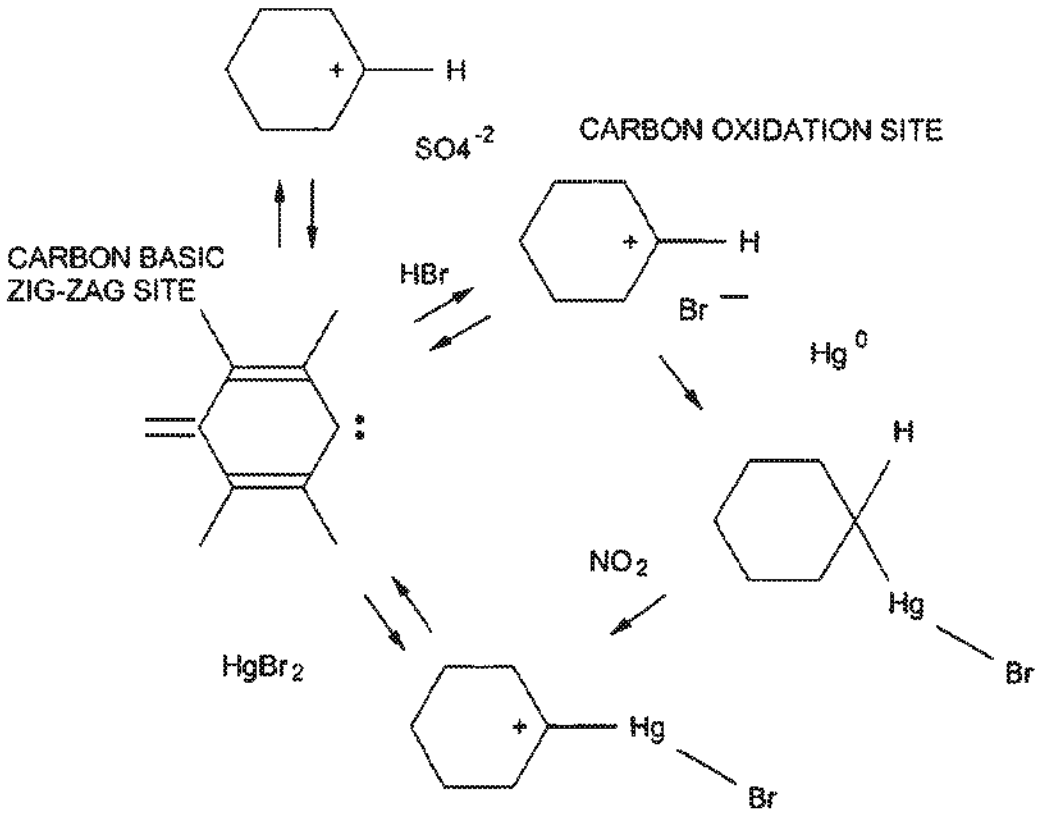


Fig. 2

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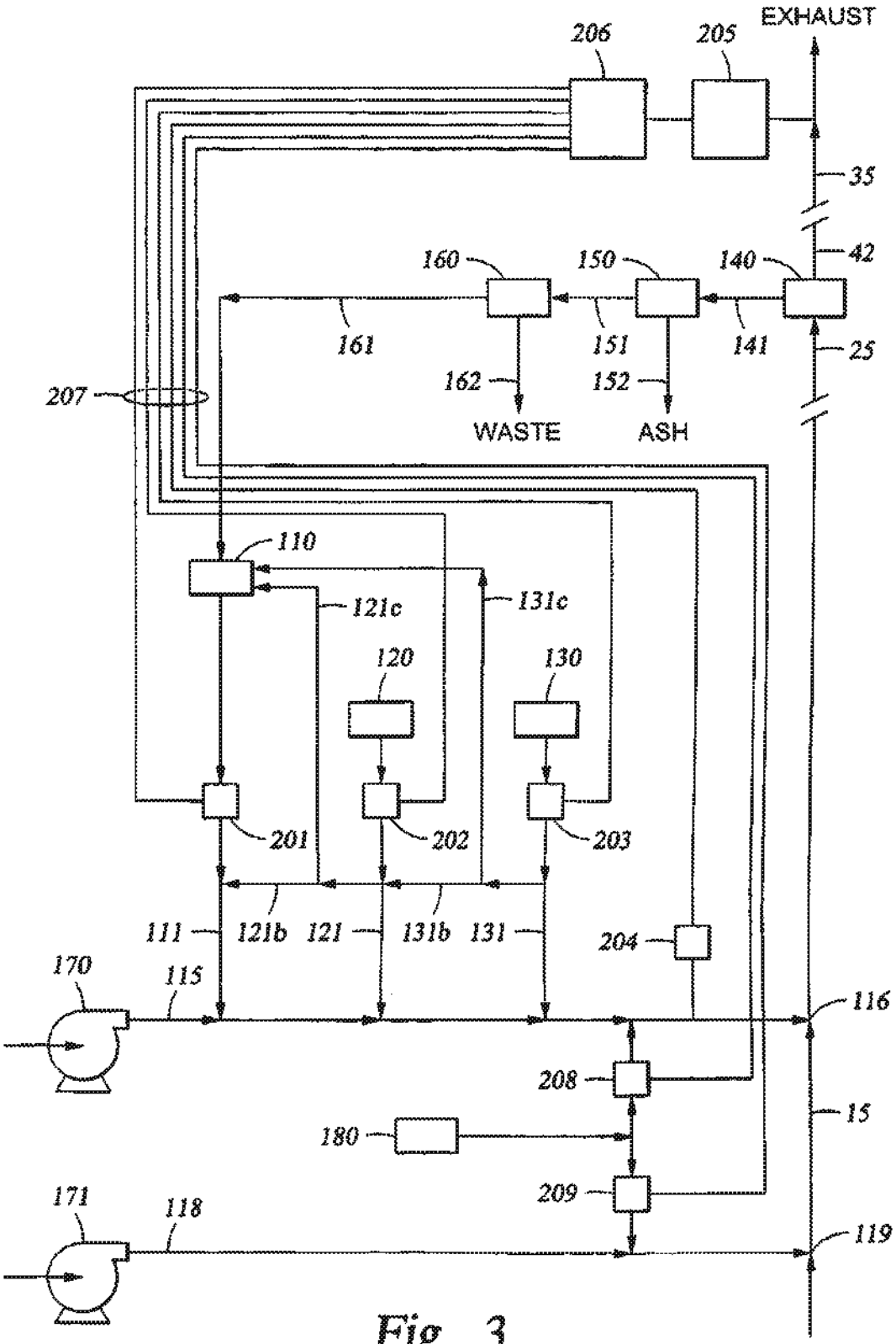

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Fig. 3

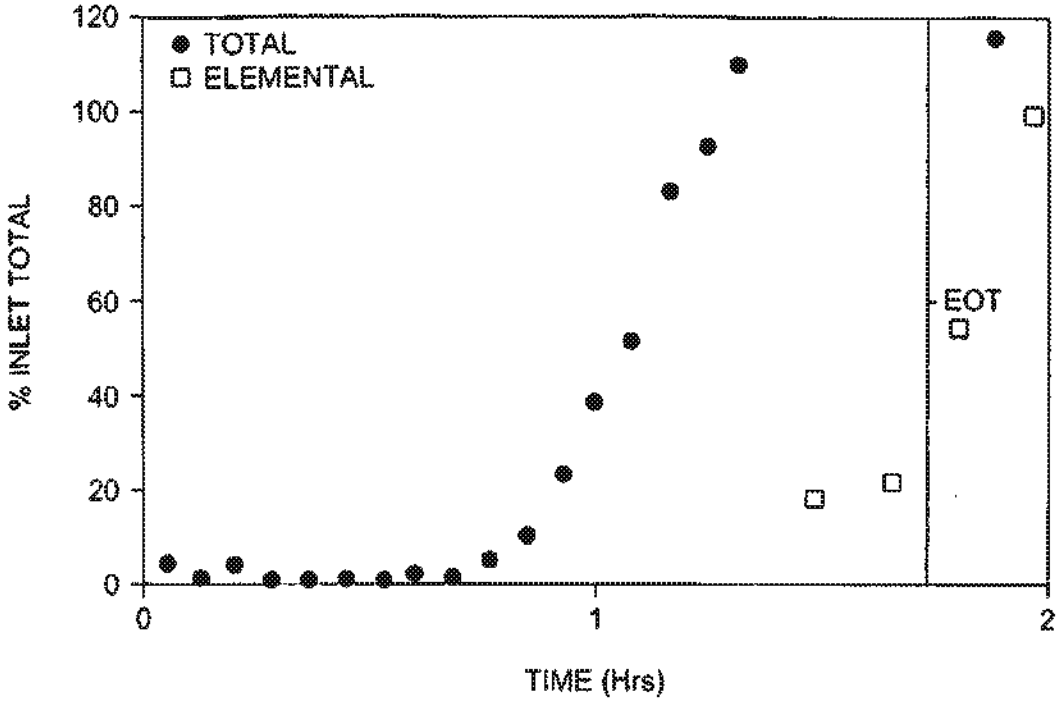


Fig. 4

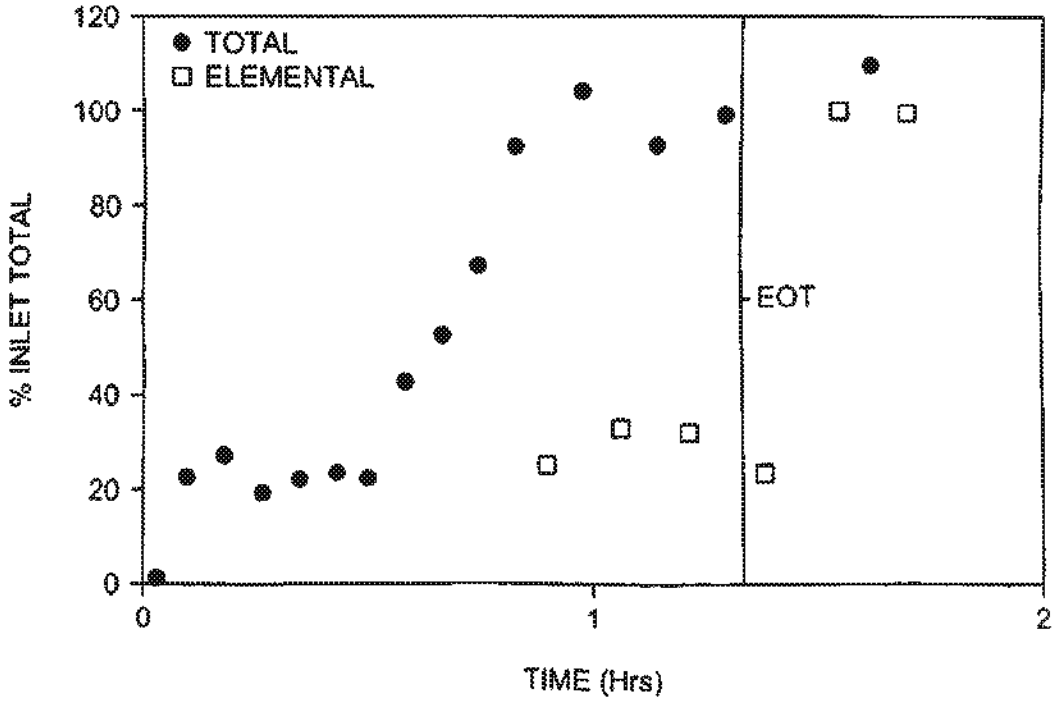


Fig. 5

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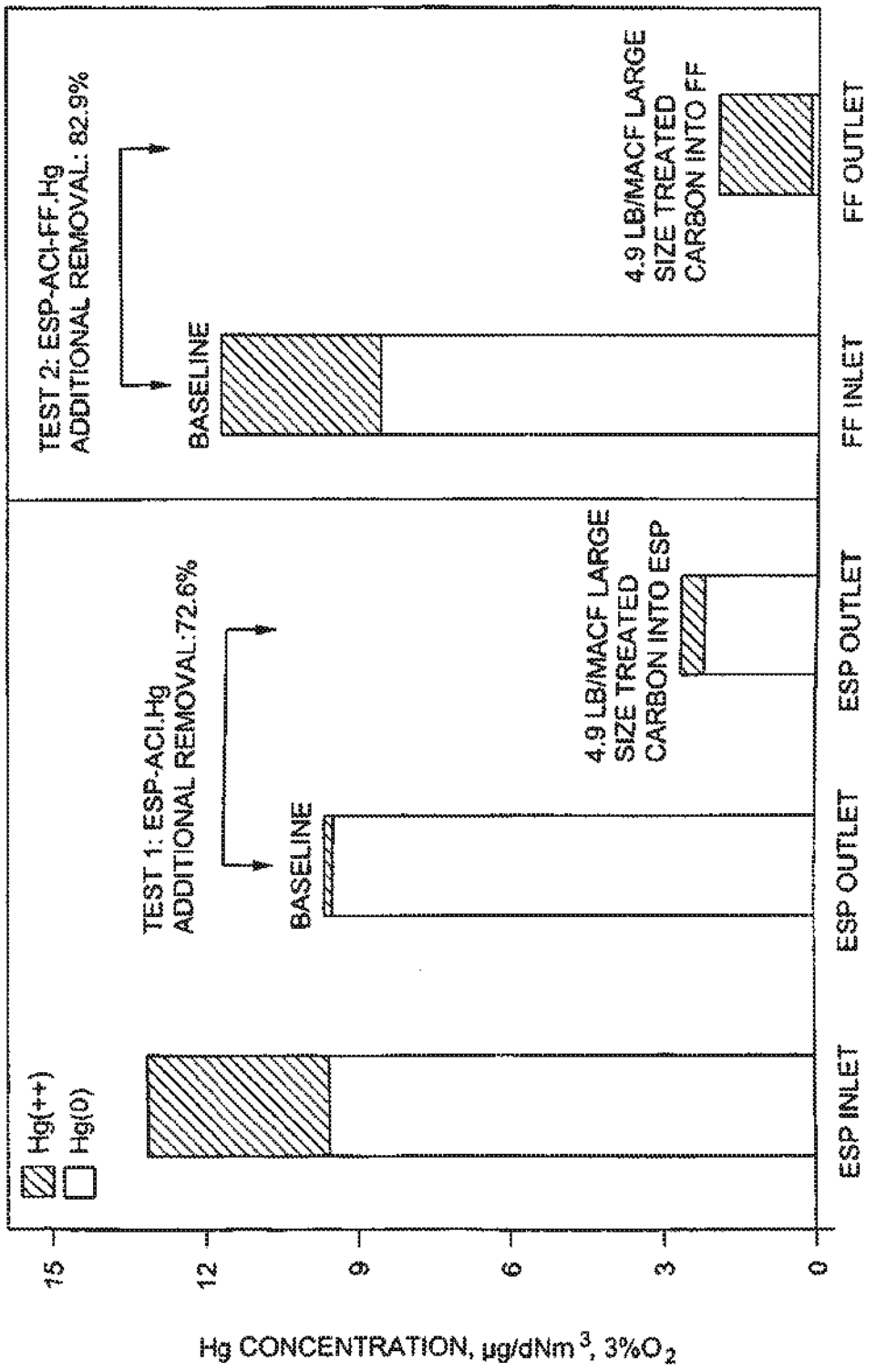


Fig. 6

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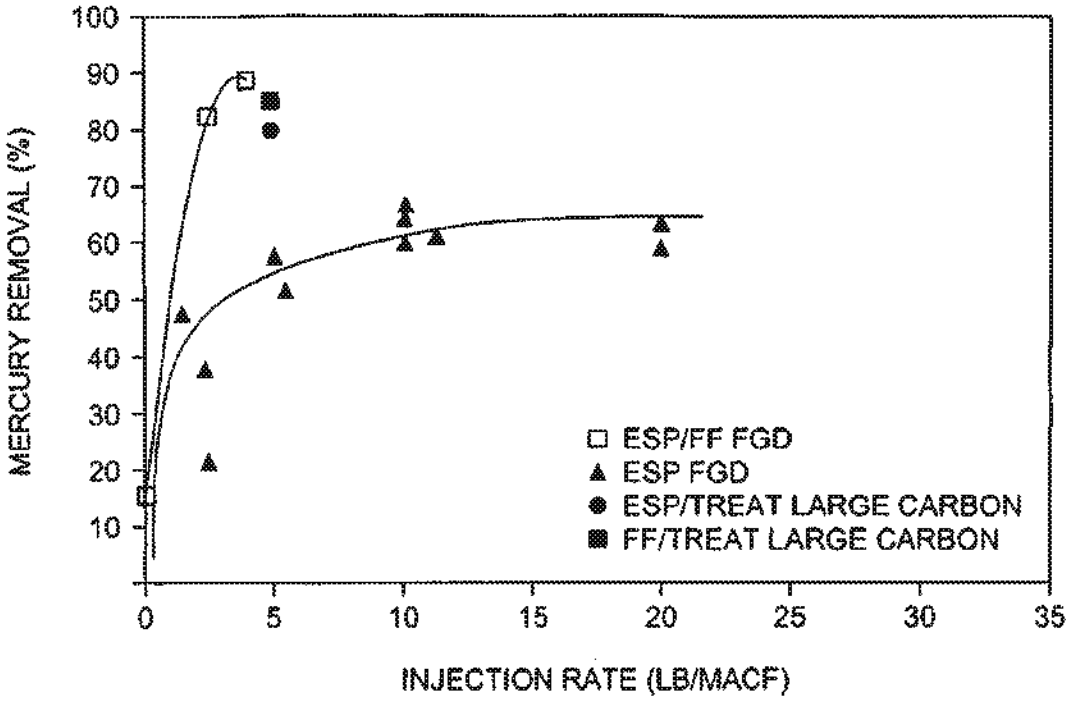


Fig. 7

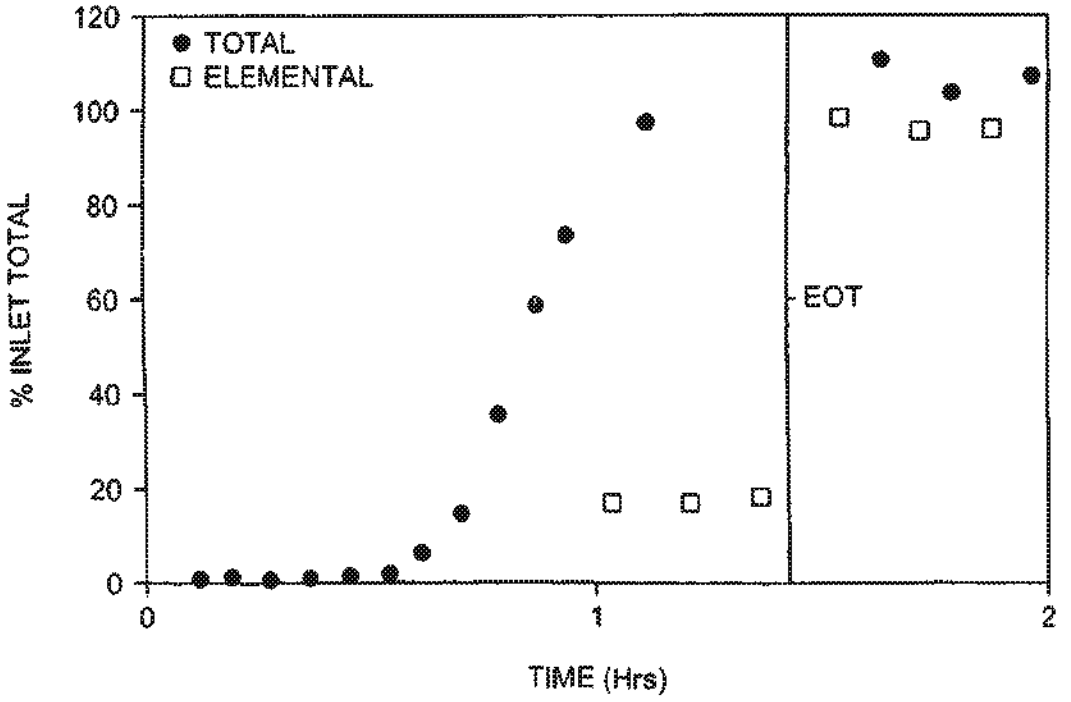


Fig. 8

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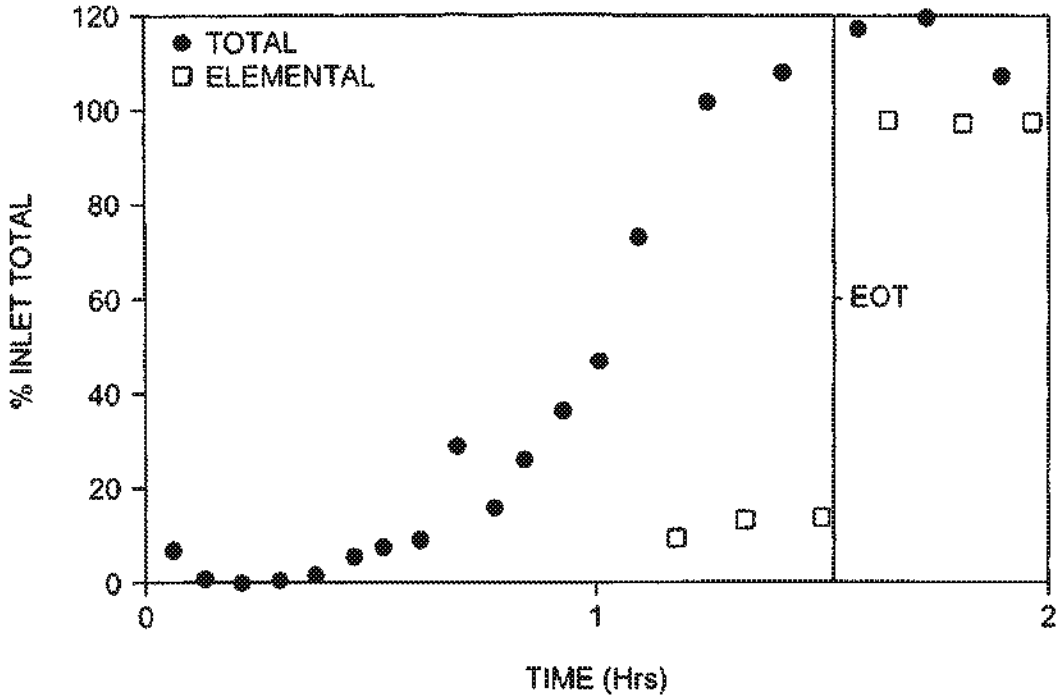


Fig. 9

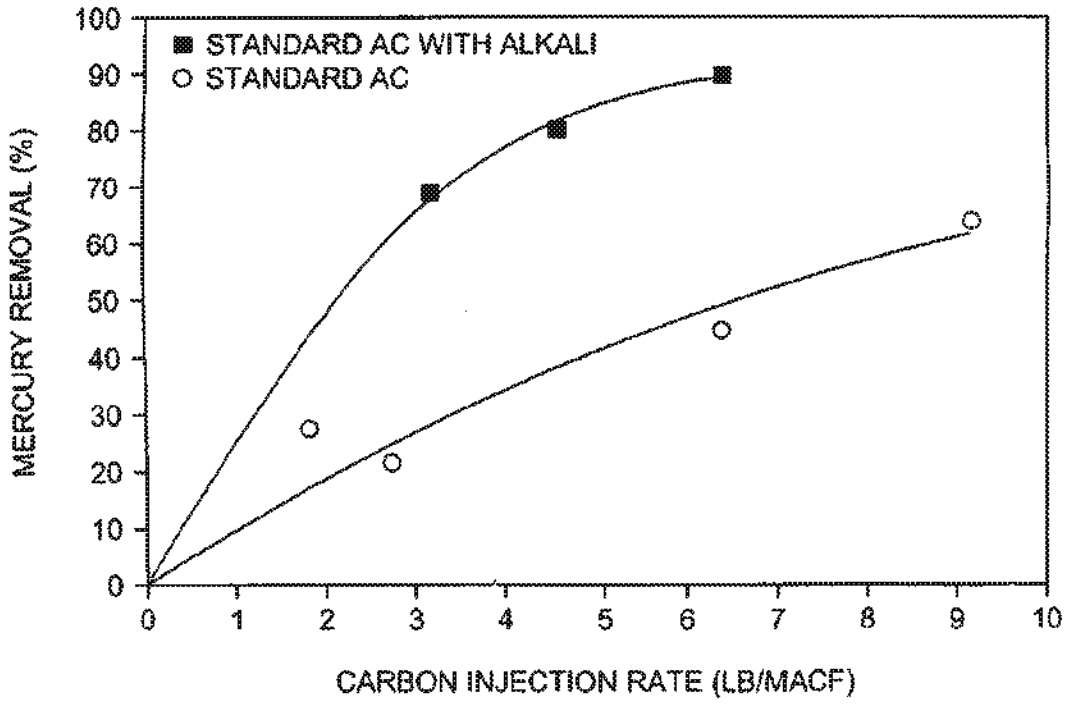


Fig. 10

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(54) Titre : SORBANTS POUR L'OXYDATION ET L'ELIMINATION DU MERCURE
(54) Title: SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

(57) Abrégé/Abstract:
A promoted carbon and/or non-carbon base sorbent are described that are highly effective for the removal of mercury from flue gas streams. The promoted sorbent comprises a carbon and/or non-carbon base sorbent that has reacted with and contains forms of halogen and halides. Optional components may be added to increase and/or preserve reactivity and mercury capacity. These may be added directly with the base sorbent, or in-flight within a gas stream (air, flue gas, etc.), to enhance base sorbent performance and/or mercury capture. Mercury removal efficiencies obtained exceed conventional methods. The promoted sorbent can be regenerated and reused. Base sorbent treatment and preparation methods are also described. New methods for in-flight preparation, introduction, and control of the active base sorbent into the mercury contaminated gas stream are described.

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(54) Title: SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

(57) Abstract: A promoted carbon and/or non-carbon base sorbent are described that are highly effective for the removal of mercury from flue gas streams. The promoted sorbent comprises a carbon and/or non-carbon base sorbent that has reacted with and contains forms of halogen and halides. Optional components may be added to increase and/or preserve reactivity and mercury capacity. These may be added directly with the base sorbent, or in-flight within a gas stream (air, flue gas, etc.), to enhance base sorbent performance and/or mercury capture. Mercury removal efficiencies obtained exceed conventional methods. The promoted sorbent can be regenerated and reused. Base sorbent treatment and preparation methods are also described. New methods for in-flight preparation, introduction, and control of the active base sorbent into the mercury contaminated gas stream are described.

SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

CROSS-REFERENCE TO RELATED APPLICATIONS

[0001] This application is being filed on 02 February 2010, as a PCT International Patent application in the name of Energy & Environmental Research Center Foundation, a U.S. national corporation, applicant for the designation of all countries except the U.S., and Edwin S. Olson, a citizen of the U.S., Michael J. Holmes, a citizen of the U.S., and John H. Pavlish, a citizen of the U.S., applicants for the designation of the U.S. only, and claims priority to U.S. Continuation-in-Part Patent Application Serial No. 12/429,058 filed on 23 April 2009.

BACKGROUND OF THE INVENTION

Technical Field of the Invention

[0002] The present invention relates to methods and materials for the removal of pollutants from flue gas or product gas from a gasification system. In particular, mercury is removed from gas streams generated during the burning or gasification of fossil fuels by highly reactive regenerable sorbents.

Background of the Invention

[0003] The combustion and gasification of fossil fuel such as coal generates flue gas that contains mercury and other trace elements that originate from the fuel. The release of the mercury (and other pollutants) to the environment must be controlled by use of sorbents, scrubbers, filters, precipitators, and other removal technologies. Mercury is initially present in the elemental form during combustion and gasification. In downstream process sections, such as in the ducts and stack of a combustion system, some of the elemental mercury is oxidized. The amount that is oxidized depends on the amount of acid gases present in the flue gas and other factors. Amounts of mercury vary with the fuel, but concentrations of mercury in the stream of flue gas from coal combustion are typically less than 5 parts per billion (ppb). Large coal combustion facilities such as electric utilities may emit a pound of mercury, or more, a day. Mercury removal applications include, without limitation, flue gas from coal (or other fossil fuel) combustion, waste incineration, product gas from gasification, as well as offgases from mineral processing, metal refining, retorting, cement manufacturing, chloralkali plants, dental facilities, and crematories.

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Mercury Sorbent Technologies

[0004] Several types of mercury control methods for flue gas have been investigated, including injection of fine sorbent particles into a flue gas duct and passing the flue gas through a sorbent bed. Fine-particle injection sorbents include activated carbon, metal oxide sorbent, sodium sulfide particles, and basic silicate or oxide sorbents. When particle injection is employed, the mercury captured on the sorbent particles is removed from the gas stream in a particulate control device such as a baghouse or electrostatic precipitator (ESP) and collected along with ash particulate. The sulfide and basic silicate and oxide particles are effective only for the oxidized mercury, and the metal oxide sorbents exhibit slower capture kinetics than the carbon particles. Additionally, injection of fine carbon particles into the flue gas stream has been only partially successful in removing mercury, especially elemental mercury, where effective removal of only about 60% is attained for some applications with a FF (fabric filter) to collect carbon and ash. Even lower removal rates have been observed when an ESP is used to collect the carbon because the contact time of the carbon with the gas is very short.

[0005] A major problem with existing carbon injection systems is that the sorbent is relatively unreactive toward mercury. Consequently, these sorbents must be used in large amounts, at high sorbent-to-mercury ratios, to effectively capture the mercury. These sorbents tend to be relatively expensive and cannot be easily separated from the ash for regeneration and reuse. The collection of carbon in the ash also creates solid waste disposal problems, and the spent sorbent may contaminate the collected ash, preventing its use in various applications.

[0006] One solution has been to add an oxidative sorbent comprising an aluminosilicate material impregnated with a very heavy dosage of one or more oxidative metal halides plus activated carbon. For example, refer to Varma et al. (20070140940). However, the amounts of metal salts required for Hg oxidation are generally relatively large and expensive. Also, several of the salts that can be used in such a process are highly toxic. Although the metal salts are present for oxidation in this process, activated carbon is essential for getting adsorption of the Hg. As such, there is no synergistic role for the aluminosilicates as they appear to be only a support for the oxidizing salts.

[0007] Another approach has been the injection of aluminosilicate particulate such as bentonite, which contains neither oxidizing salts nor halogen complexes with a Lewis base site, and thus lacks the more powerful oxidizing capability of the said complexes as described in this application. For example, see U.S. Patent No. 7,413,719. Additionally, the injection of an aluminosilicate (kaolin or metakaolin) containing

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calcium hypochlorite which thermally decomposes to form halogen is also known. For example, see U.S. Patent Application No. 20030103882. Thus these and similar impregnated aluminosilicate technologies require time in flight at appropriate high temperatures to heat the impregnated salt(s) to generate an oxidation site. This clearly represents a kinetic barrier to activation in contrast to the extremely fast complexing reaction of the Lewis acid on the surface of the appropriate Lewis base sorbent described in the present patent. The kinetic barrier is only for heating up the calcium hypochlorite to decompose it to Cl atoms or molecules. Halogen (bimolecular or atomic) would complex with carbon or noncarbon at any lower temperature to form reactive oxidation sites. Also, halide would require a very high temp or strong acid to form reactive halogen or halogen complex.

[0008] Yet another approach is the injection of bentonite plus a metal sulfide and a metal salt, none of which is oxidizing to elemental mercury and would require a slow thermal activation step. For example, see U.S. Patent Application No. 20070119300.

[0009] The injection of halogen or halogen precursors in a hot zone, followed by contact with an alkaline material in a wet or dry scrubber is another approach known in the art. With such an approach, elemental mercury is claimed to be oxidized by the halogen to Hg(II) which is collected by the alkaline material in the scrubber. For example, see U.S. Patent No 6,808,692 (Oehr), U.S. Patent No. 3,849,267 (Hilgen), U.S. Patent No. 5,435,980 (Felsvang), U.S. Patent No. 6,375,909 (Dangtran), U.S. Patent Application No. 20020114749 (Cole), U.S. Patent No. 6,638,485 (Iida), U.S. Patent Application No. 20030185718 (Sellakumar), U.S. Patent Application No. 20030147793 (Breen), and U.S. Patent No. 6,878,358 (Vosteen). However, even though it is known to inject halogen forms at some stage of the combustion process, such a process does not utilize a complexing method on a sorbent surface for conducting the oxidation and capture. Further, the alkaline material is rapidly surface-coated by the large concentrations of acid gases, lowering its capacity for adsorption of Hg(II). It is also recognized that the halogen forms initially introduced or generated are far more reactive to the large concentrations of SO_2 and moisture in the flue gas, and so gas-phase reactions of the halogens with Hg are hindered. In contrast, the present invention takes advantage of the Lewis acid complexes that rapidly form on the sorbent surface to effect the Hg oxidation, rather than rely on gas phase reactions for oxidation. Thus HCl, HBr, SO_2Br, and other gas-phase products all festoon the surface and promote the activity of the sorbent by forming complexes with the sorbent to form a promoted sorbent.

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[0010] Accordingly, there remains a need for more economical and effective mercury removal technology. This invention provides for cost-effective removal of pollutants, including mercury, using sorbent enhancement additives and/or highly reactive sorbents, with contact times of seconds (or less), and that may be regenerated and reused.

SUMMARY

[0011] The various embodiments of the present invention overcome the various aspects of the deficiencies of the prior art and provide new and economical methods for the removal of mercury from the gases produced in the utilization of fossil fuels.

[0012] A halogen/halide-promoted sorbent is described that is highly effective for the removal of mercury from flue gas streams. The sorbent comprises any activated carbon and/or non-carbon compound, such as porous or vesicular felsic or basaltic materials, clay-based compounds, alkaline compounds, calcium hydroxide compounds, sodium acetate compounds, and/or bicarbonate compounds, or a combination thereof. Optional secondary components and alkali may be added to further increase reactivity and mercury capacity. Mercury removal efficiencies obtained exceed or match conventional methods with added benefits such as reduced costs. Optionally, the promoted sorbent can be regenerated and reused. Sorbent treatment and/or preparation methods are also described. Methods for in-flight preparation, introduction, and control of the sorbent, promoter and promoted sorbent into the mercury contaminated gas stream are described.

[0013] When a promoted or a non-promoted base sorbent reacts with elemental or oxidized mercury, a mercury/sorbent chemical composition is formed and, in the case of elemental mercury reacting with the promoted base sorbent, the mercury is oxidized. As discussed in further detail in other portions of the specification, the base sorbent may be either a carbon or a non-carbon material or a combination thereof. Additionally, the mercury/sorbent chemical composition may be comprised of covalent bonds, ionic bonds and/or chemical complexes between the promoted or non-promoted base sorbent and the oxidized mercury. The Lewis basic groups on the non-promoted base sorbent, or the non-promoted portions of a promoted base sorbent, are available for reaction with the Lewis acid groups of already existing oxidized mercury in the mercury containing gas. Thus, mercury may be removed from the mercury containing gas stream through the formation of multiple and various mercury/sorbent chemical compositions even within the same process. For example, multiple sites on a sorbent particulate can form

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multiple and various mercury/sorbent chemical compositions in the case where only a portion of the sites on the base sorbent particulate are promoted.

[0014] In some embodiments, a carbon and/or non-carbon promoted sorbent and/or a combination thereof is provided comprising a sorbent structure that has reacted with a promoter selected from the group consisting of halides, halogens, and combinations thereof, such that the reaction product is effective for the removal of mercury from a gas stream. The carbon sorbent comprises reactive Lewis acid groups/sites; the non-carbon sorbent comprises reactive Lewis basic groups/sites.

[0015] In an embodiment, a promoted carbon and/or non-carbon sorbent is provided wherein the base sorbent is selected from the group consisting of carbon, activated carbon, porous or vesicular felsic and basaltic materials, clay-based compounds, alkaline compounds, calcium hydroxide compounds, sodium acetate compounds, and/or bicarbonate compounds, or a combination thereof, with an average particle size similar to that of fly ash produced from a thermal process (combustion or gasification) or greater than that of fly ash produced such that it is physically separable therefrom, and combinations thereof, and the promoter is selected from the group consisting of molecular halogens, Group V (CAS nomenclature is used throughout) halides, Group VI halides, hydrohalides, and combinations thereof. In an embodiment, the promoted sorbent (carbon, non-carbon, or their combination) may have a mass mean particle diameter such that it can be substantially separated by physical means from entrained ash in the gas stream from which mercury is to be removed. In an embodiment, the base sorbent (carbon, non-carbon, or their combination) may have a mass mean particle diameter greater than about 40 micrometers.

[0016] In another embodiment, the promoted sorbent comprises from about 1 to about 30 grams of promoter per 100 grams of base sorbent. Another embodiment further comprises an optional secondary component comprising a halogen or a hydrohalide such that the reactivity and mercury capacity of the promoted sorbent are enhanced.

[0017] In another embodiment, the concentration of the optional secondary component on the finished sorbent is within the range of from about 1 to about 15 wt% of the concentration of the promoter on the finished sorbent.

[0018] In another embodiment, an optional alkali component may preferably be added to provide a synergistic effect through combination of this alkali with the base sorbent.

[0019] In another embodiment, the optional secondary component is selected from the group consisting of Group V halides, Group VI halides, HI, HBr, HCl, and combinations thereof. In another embodiment, the promoter is substantially in vapor

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form when combined with the base sorbent. In another embodiment, the promoter is combined with an organic solvent prior to reaction with the base sorbent. In another embodiment, the promoter and optional secondary component are combined with the base sorbentsubstantially simultaneously. Another embodiment further comprises adding a mercury-stabilizing reagent selected from the group consisting of S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof. Another embodiment further comprises adding an optional alkali component.

[0020] In an embodiment, a method is provided comprising providing a granular base sorbent and reacting the base sorbent with a promoter selected from the group consisting of halogens, halides, and combinations thereof, such that the reaction product comprises a promoted sorbent effective for removal of mercury from a gas stream. In a further embodiment, the reaction product comprises from about 1 to about 30 grams of promoter per 100 grams of base sorbent. In another embodiment the reaction product has an average particle-size distribution greater than the average size of entrained ash particles in the gas stream from which mercury is to be removed, such that the reaction product can be substantially removed from the entrained ash particles by physical means. In another embodiment, the reaction product has a mass mean particle diameter greater than about 40 micrometers.

[0021] In another embodiment, the promoter is selected from the group consisting of molecular halogens, hydrohalides, Group V halides, Group VI halides, and combinations thereof. In another embodiment, the promoter is in the gas phase when contacting the base sorbent (carbon, non-carbon, or their combination). In another embodiment, the promoter is in an organic solvent when contacting the base sorbent (carbon, non-carbon, or their combination).

[0022] In another embodiment, the promoter is selected from the group consisting of Br_2, a Group V bromide, a Group VI bromide, and combinations thereof.

[0023] In another embodiment, the method further comprises reacting the granular non-carbon with an optional secondary component comprising a halogen or a hydrohalide such that the reactivity and mercury capacity of the promoted sorbent are enhanced. In another embodiment, the promoter and optional secondary component are contacted simultaneously with the non-carbon base sorbent. In another embodiment, the method further comprises adding a mercury-stabilizing reagent selected from the group consisting of S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof. In an embodiment, a method is provided for control of mercury in a flue gas with substantially lower sorbent requirements. Through enhanced sorbent reactivity,

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mercury removal per gram of sorbent is increased, thereby decreasing the capital and operating costs by decreasing sorbent requirements.

[0024] In an embodiment, the promoted sorbent is introduced by direct injection into the flue gas stream. In another embodiment, the base sorbent is promoted within the flue gas stream

[0025] In an embodiment, a method is provided for reducing mercury in flue gas comprising providing a base sorbent, either by injection or in situ creation, into a mercury-containing flue gas stream, collecting greater than 70 wt% of the mercury in the flue gas on the promoted sorbent to produce a cleaned flue gas, and substantially recovering the promoted sorbent from the cleaned flue gas. In embodiments where less than 70 wt% mercury removal is desired, the required removal is attained using less base sorbent as would be required with standard base sorbent. In a further embodiment, the method further comprises monitoring the mercury content of the clean flue gas, regenerating the recovered promoted sorbent, and using the monitored mercury content of the cleaned flue gas to control the rate of base sorbent and promoter. In another embodiment the injected promoted sorbent is prepared in-flight by reacting a base sorbent (carbon, non-carbon, or their combination) and a promoter within a pneumatic transport line from which the reaction product is injected to the mercury-containing flue gas stream.

[0026] In another embodiment, the promoter is selected from the group consisting of molecular halogens, halides, and combinations thereof. In another embodiment, the promoter is reacted in the gas phase or as a vapor. In another embodiment, the promoter is added at from about 1 to about 30 grams per 100 grams of the base sorbent (carbon, non-carbon, or their combination).

[0027] In another embodiment, the injected promoted sorbent is prepared in-flight by reacting a base sorbent (carbon, non-carbon, or their combination), a promoter, and an optional secondary component to enhance the reactivity and capacity of the promoted sorbent within a pneumatic transport line from which the reaction product is injected to the mercury-containing flue gas stream.

[0028] In another embodiment, the optional secondary component is selected from the group consisting of iodine, hydrohalides, Group V halides, Group VI halides, and combinations thereof. In another embodiment, the optional secondary component is added at from about 1 to about 15 wt% of the promoter content. In another embodiment, the method further comprises adding to the promoted sorbent a mercury-stabilizing reagent selected from the group consisting of S, Se, H_2S, SO_2, H_2Se, SeO_2,

CS_2, P_2S_5, and combinations thereof.

[0029] In an embodiment, the method further comprises coinjecting an optional alkaline material, including without limitation alkaline and alkaline-earth components, to improve the efficiency of mercury capture by capturing oxidized mercury and/or capturing gaseous components that might otherwise reduce promoted sorbent capacity. In another embodiment, the optional alkaline material may preferably comprise calcium oxide, sodium carbonate, and the like, as are known in the art.

[0030] In another embodiment, the method further comprises using the monitored mercury content of the cleaned flue gas to control the composition of the promoted sorbent. In another embodiment, the promoted sorbent is prepared in-flight by reacting a base sorbent (carbon, non-carbon, or their combination) and a promoter within the flue gas stream or in a transport line from which the reaction product is injected to the mercury-containing flue gas stream, wherein the promoter is selected from the group consisting of molecular halogens, halides, and combinations thereof, wherein the promoter is reacted in the gas phase or as a vapor, wherein the promoter is added at from about 1 to about 30 grams per 100 grams of the base sorbent (carbon, non-carbon, or their combination), wherein the rate at which the promoter is added and the rate of promoted sorbent injection are determined by a digital computer based, at least in part, on the monitored mercury content of the cleaned flue gas.

[0031] In an embodiment, a method for reducing the mercury content of a mercury and ash-containing gas stream is provided wherein particulate carbon and/or non-carbon promoted sorbent with a mass mean size greater than 40 μm is injected into the gas stream, mercury is removed from the gas by the promoted sorbent particles, the promoted sorbent particles are separated from the ash particles on the basis of size, and the promoted sorbent particles are reinjected to the gas stream. In another embodiment, the mercury-containing promoted sorbent particles are regenerated to remove some or substantially all of the mercury. In another embodiment, an alkaline component is coinjected into the gas stream. In another embodiment, the promoted sorbent may further comprise a promoter. The promoter may preferably comprise a halide, a halogen, or both.

[0032] In an embodiment, a method for reducing mercury in a mercury-containing gas to a desired level is disclosed. The method comprises reacting a carbon base sorbent with at least one promoter selected from the group consisting of molecular halogens, halides, and combinations thereof to produce a promoted carbon sorbent; allowing said promoted carbon sorbent to interact with a mercury-containing gas to capture mercury

in the mercury-containing gas on the promoted sorbent to produce a cleaned gas; and monitoring the mercury content of the cleaned gas. In some embodiments, the carbon base sorbent and the promoter are introduced into the mercury-containing gas at the same location or at separate locations. In some embodiments, the carbon base sorbent or promoter or combination thereof is introduced into the mercury-containing gas at one or more locations. In an embodiment, the rate at which the carbon base sorbent is introduced or the rate at which the promoter is introduced or combination thereof is adjusted according to the monitored mercury content of the cleaned gas so that the mercury content of the cleaned gas is maintained at substantially the desired level with minimal operating cost.

[0033] In a further emobodiment, the method comprises reacting a non-carbon base sorbent with at least one promoter selected from the group consisting of molecular halogens, halides, and combinations thereof to produce a promoted non-carbon sorbent; allowing said promoted non-carbon sorbent to interact with a mercury-containing gas to capture mercury in the mercury-containing gas on the promoted sorbent to produce a cleaned gas; and monitoring the mercury content of the cleaned gas. In some embodiments, the non-carbon base sorbent and the promoter are introduced into the mercury-containing gas at the same location or at separate locations. In some embodiments, the non-carbon base sorbent or promoter or combination thereof is introduced into the mercury-containing gas at one or more locations. In an embodiment, the rate at which the non-carbon base sorbent is introduced or the rate at which the promoter is introduced or combination thereof is adjusted according to the monitored mercury content of the cleaned gas so that the mercury content of the cleaned gas is maintained at substantially the desired level with minimal operating cost.

[0034] In a further embodiment, a method for reducing mercury in a mercury-containing gas to a desired level is presented. The method comprises reacting a base sorbent with at least one promoter selected from the group consisting of molecular halogens, halides, and combinations thereof to produce a promoted sorbent, wherein said base sorbent is selected from the group consisting of a non-carbon material, a carbon material, and combination thereof; allowing said promoted sorbent to interact with a mercury-containing gas to capture mercury in the mercury-containing gas on the promoted sorbent to produce a cleaned gas; and monitoring the mercury content of the cleaned gas.

[0035] In some embodiments, the base sorbent and the promoter are introduced into the mercury-containing gas at the same location or at separate locations. In some

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embodiments, the base sorbent or promoter or combination thereof is introduced into the mercury-containing gas at one or more locations. In some embodiments, introducing the base sorbent and the promoter comprises injecting the base sorbent and the promoter into the mercury-containing gas. In some embodiments, the promoter is introduced into the mercury-containing gas upstream of the introduction of the base sorbent. In some embodiments, the promoter is introduced upstream of a boiler or a combustion chamber. In an embodiment, the rate at which the base sorbent is introduced or the rate at which the promoter is introduced or combination thereof is adjusted according to the monitored mercury content of the cleaned gas so that the mercury content of the cleaned gas is maintained at substantially the desired level with minimal operating cost.

[0036] In an embodiment, the base sorbent for the promoted sorbent is selected from the group consisting of carbon, activated carbon, porous felsic materials, vesicular felsic materials, porous basaltic materials, vesicular basaltic materials, clay-based compounds, alkaline compounds, calcium hydroxide compounds, sodium acetate compounds, bicarbonate compounds, and combinations thereof. In embodiments, the non-carbon material comprises Lewis basic groups and the carbon material comprises Lewis acid groups. In some cases, the non-carbon material comprises amorphous forms of tectosilicates comprising nanoscale cavities lined with Lewis basic oxygen associated with alkaline-earth metals. The alkaline-earth metals comprise Group I and Group II alkaline-earth metals. In some other cases, the non-carbon material comprises amorphous forms of phyllosilicates comprising nanoscale cavities lined with Lewis basic oxygen.

[0037] In an embodiment, the promoted sorbent comprises metastable complexes formed between the promoter of this disclosure and inorganic species on the non-carbon base sorbent. In some embodiments, the inorganic species is selected from the group consisting of sodium compounds, calcium compounds, magnesium compounds, aluminum compounds, iron compounds, and combinations thereof. In an embodiment, the promoted sorbent comprises metastable complexes formed between the promoter of this disclosure and metal-oxygen-metal structures on the non-carbon base sorbent. In some embodiments, the promoter after being complexed with the metal-oxygen-metal structures is in the form selected from the group consisting of a dihalogen group, a halogen atom, a hydrohalogen group, a Group V halide, a Group VI halide, and combinations thereof. In an embodiment, the promoted sorbent comprises activated Lewis basic groups or activated Lewis acid groups or combination thereof. In some embodiments, the interaction between promoted sorbent and said mercury-containing

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gas stream comprises mercury diffusing from the gas phase onto said promoted sorbent surface; and reacting with the activated Lewis basic groups or activated Lewis acid groups or combination thereof to cause chemisorption on the promoted sorbent surface.

[0038] In an embodiment, the method for reducing mercury in a mercury-containing gas to a desired level further comprises pretreating the base sorbent to increase the number of Lewis basic groups or Lewis acid groups or combination thereof. In some cases, the pretreating methods comprise chemical treatment, thermal treatment, vacuum treatment, and combinations thereof. In some embodiments, chemical treatment comprises acid treatment and alkaline treatment. In an embodiment, [[,]] the method for reducing mercury in a mercury-containing gas to a desired level further comprises introducing an alkali component into the mercury-containing gas.

[0039] As will be described in more detail below, the present invention thus provides several advantages over previously known techniques, including significantly more effective and economical mercury sorbents for effluent gases, advantageously applicable to treating gas streams from combustion and gasification systems.

[0040] The foregoing has outlined rather broadly the features and technical advantages of the present invention in order that the detailed description of the invention that follows may be better understood. Additional features and advantages of the invention will be described hereinafter that form the subject of the claims of the invention. It should be appreciated by those skilled in the art that the conception and specific embodiments disclosed may be readily utilized as a basis for modifying or designing other structures for carrying out the same purposes of the present invention. It should also be realized by those skilled in the art that such equivalent constructions do not depart from the spirit and scope of the invention as set forth in the appended claims.

BRIEF DESCRIPTION OF THE DRAWINGS

[0041] For a more detailed description of the preferred embodiments of the present invention, reference will now be made to the accompanying drawings.

[0042] Figure 1 schematically illustrates methods for preparation of promoted carbon and/or non-carbon sorbents in accordance with the present invention.

[0043] Figure 2 illustrates a proposed mechanistic model of the chemical reactions resulting in the oxidation and capture of mercury.

[0044] Figure 3 schematically illustrates preparation of promoted carbon and/or non-carbon sorbents and processes for flue gas mercury reduction in flue gases and/or

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product gases from a gasification system in accordance with the present invention, including in-flight preparation of promoted carbon and/or non-carbon sorbent.

[0045] Figure 4 illustrates a mechanism for promotion of metal oxide base sorbent via formation of a reactive halogen complex sorbent and subsequent capture of elemental mercury on the promoted sorbent.

[0046] Figure 5A schematically illustrates an exemplary process flow diagram for in-flight preparation of a promoted carbon and/or non-carbon sorbent.

[0047] Figure 5B schematically illustrates an exemplary process flow diagram for in-flight preparation of a promoted carbon and/or non-carbon sorbent.

DETAILED DESCRIPTION

[0048] Herein will be described in detail specific preferred embodiments of the present invention, with the understanding that the present disclosure is to be considered an exemplification of the principles of the invention and is not intended to limit the invention to that illustrated and described herein. The present invention is susceptible to preferred embodiments of different forms or order and should not be interpreted to be limited to the specifically expressed methods or compositions contained herein. In particular, various preferred embodiments of the present invention provide a number of different configurations and applications of the inventive method, compositions, and their uses.

[0049] The present invention provides a cost-effective way to capture pollutants by utilizing exceptionally reactive halogen/halide-promoted sorbents using a bromide (or other halogen/halide) treatment of the promoted sorbent, that capture mercury via mercury–sorbent surface reactions, at very short contact times of seconds or less. The reactivity of the promoted sorbent toward the pollutants (i.e., mercury) is greatly enhanced, and the sorption capacity can be regenerated; i.e., the promoted sorbent may be regenerated, recycled and/or reused.

[0050] The treated base sorbents (carbon, non-carbon, or their combination), treatment techniques, and optional additives discussed herein have applicability to mercury control from the product or effluent gas or gases from gasification systems, syngas generators, and other mercury-containing gas streams, in addition to the flue gas from combustion systems. Thus it should be understood that the terms combustion system and flue gas as used throughout this description may apply equally to gasification systems and syngas or fuel gas, as will be understood by those skilled in the art.

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[0051] Hereinafter the disclosure may at times discuss the use of carbon base sorbents in further details; however the use of non-carbon base sorbents and a combination of carbon and non-carbon base sorbents is also contemplated to at least the same degree as carbon base sorbents.

[0052] Referring now to Figure 1, there is shown a block flow diagram illustrating some preferred embodiments of the process of the present invention to prepare promoted sorbents useful for mercury capture in a mercury containing gas, such as a flue gas and/or product gas from gasification system streams. In a preferred embodiment illustrated by path 10-20, block 10 illustrates providing a base sorbent while block 20 illustrates adding a halogen or halide promoter that reacts with the base sorbent to produce a product-promoted sorbent. In embodiments where the halogen or halide is added, for example, as a vapor, no further steps may be necessary. In embodiments where the halogen or halide is added in, for example, a solvent, it may be desirable to employ solvent removal as illustrated by block 20A to produce a product-promoted sorbent suitable for injection.

[0053] Referring still to Figure 1, another preferred embodiment of the process of the present invention is illustrated by path 10-20-30, comprising providing a base sorbent as shown by block 10, adding a halogen or halide promoter that reacts with the base sorbent, illustrated at block 20, and adding a secondary component illustrated at block 30 that reacts with the result of block 20 to produce a product-promoted sorbent. In embodiments where both the halogen or halide promoter and the secondary component are added, for example, as a vapor, no further steps may be necessary. In embodiments where the halogen or halide promoter and/or secondary component are added in, for example, a solvent, it may be desirable to employ solvent removal as illustrated by block 30A to produce a product-promoted sorbent suitable for injection.

[0054] Referring still to Figure 1, another preferred embodiment of the process of the present invention is illustrated by path 10-40, comprising providing a base sorbent as illustrated at block 10 and adding a halogen or halide promoter and a secondary component to the base sorbent together, with which they react to produce a product-promoted sorbent as illustrated by block 40. As above, in embodiments where vapor additions are made to the base sorbent, no further steps may be desired. In embodiments where one or more components are added in solvent, a solvent removal step may be provided as illustrated by block 40A to produce a product-promoted sorbent suitable for injection.

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[0055] Referring still to Figure 1, another preferred embodiment of the process of the present invention is illustrated by path 10-50 in combination with path 20-50. In this embodiment, a base sorbent as illustrated by block 10 is introduced to the mercury containing gas as illustrated by block 50 while a halogen or halide promoter as illustrated by block 20 is introduced to the mercury containing gas stream. Thus the base sorbent and promoter react at block 50 to produce a product-promoted sorbent. In a similar manner, a secondary component as illustrated by block 30 may be added to the halogen or halide promoter as illustrated by block 20 and introduced into the mercury containing gas as illustrated by block 50. In embodiments where both the halogen or halide promoter and secondary component are added, for example, by vapor, no further steps may be taken. In embodiments where the halogen or halide and/or secondary component are added in, for example, a solvent, it may be desirable to employ solvent removal as illustrated by block 20A and/or block 30A.

[0056] Referring still to Figure 1, also illustrated are preferred embodiments in which, as illustrated by block 50, a mercury containing gas stream is treated with product-promoted carbon sorbent prepared as described above.

[0057] In some preferred embodiments, the carbon base sorbent provided may preferably be any of several types, as understood by those skilled in the art. For example, the carbon base sorbent may include powdered activated carbon, granular activated carbon, carbon black, unburned carbon, carbon fiber, carbon honeycomb or plate structure, aerogel carbon film, pyrolysis char, regenerated activated carbon from product-promoted carbon sorbent, or other types as known in the art.

[0058] In some preferred embodiments, the carbon base sorbent provided may preferably have a mass mean particle size greater than the fly ash in a mercury containing gas, such as a flue gas stream, to be treated.

[0059] In some preferred embodiments, the carbon base sorbent provided may preferably have a mass mean particle diameter preferably greater than 40 micrometers, more preferably greater than 60 micrometers, or a particle-size distribution greater than that of the fly ash or entrained ash in a flue gas stream, or other mercury containing gas, to be treated, such that the activated carbon and ash can be separated by physical means.

[0060] In some preferred embodiments, the halogen or halide promoter that is added to, and reacts with, the carbon base sorbent may preferably comprise, by way of illustration and not limitation, a molecular halogen in vapor or gaseous form, a molecular halogen in an organic solvent, a Group V or Group VI halide, such as PBr_3 or SCl_2, respectively, in vapor, liquid, or solution form (though not in an aqueous solvent).

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[0061] Embodiments are also provided in which the organic solvent may preferably comprise a chlorinated hydrocarbon, such as dichloromethane, a hydrocarbon solvent, including for example, petroleum ether, ligroin, pentane, hexane, toluene, and benzene, carbon disulfide, a waste solvent, an ether, a recycled solvent, a supercritical solvent, such as supercritical CO_2, water (though not in the case of a Group V or Group VI halide), and others as will be apparent to those of skill in the art.

[0062] Referring now to Figure 4, a theory is illustrated developed from scientific evidence to explain the nature of the promoting compounds. For example, as illustrated in Figure 4, a molecular bromine molecule forms a complex with the surface of the base sorbent comprising a high surface area form of a metal oxide. Complexing with bromine can occur at surface and defect sites on the surface of the glassy amorphous tectosilicates by association with the Lewis basic oxygen lining the interstitial cavities and the alkali actions on the surface. Molecular hydrogen bromide or an electrophilic or Lewis acid bromine compound react to form a similar structure. The precedence for this promotion of a metal oxide surface complex with an acidic species is described in a paper by Stark and Klabunde (Klabunde, K.J. Chem. Mater. 1996, 8, 1913-1918) who showed addition of acids, HCl, SO_3, and NO, to a MgO surface to form a surface complex. In US Patent 6,517,423 to Koper et al. the described surface complexes were active for destroying biological agents and toxins. As shown in Figure 4, addition of halogens to a metal oxide surface (A) can form a complex that could be described as a bromide-oxybromide species (B), in which electrophilic reactivity is maintained, owing to the formation of positive charges on the surface. The electrophilic complex formed on the metal oxide base sorbent comprises an active site for oxidation of elemental mercury. Addition of mercury to the complex results in formation of a mercury oxygen bond and, simultaneously, a mercury bromine bond as shown in complex C in Figure 4. Thus the final structure is a stable oxidized Hg form (D) described as a metaloxymercuric bromide.

[0063] In summary, it has now been found that the formation of the new bromide complex with the metal oxide surface increases the surface reactivity toward mercury and other pollutants. Additionally, the resulting bromide compound is uniquely suited to facilitate oxidation of the mercury. The effectiveness of the oxidation apparently results from the promotion effect of the halide, exerted on the developing positive charge on the mercury during the oxidation, known in the chemical art as a specific catalytic effect. Thus as the mercury electrons are drawn toward the positive surface oxygen, the halide anion electrons are pushing in from the other side, stabilizing the

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positive charge developing on the mercury and lowering the energy requirement for the oxidation process. Bromide is especially reactive, owing to the highly polarizable electrons in the outer 4p orbitals of the ion. Thus adding HBr or Br_2 to the appropriate metal oxide forms a similar surface bromide complex, in which the positive oxygen oxidizes the mercury with the assistance of the bromide ion.

[0064] In embodiments, a non-carbon base sorbent with Lewis basic sites/groups comprising metal-oxygen-metal structures is activated by a promoter of this disclosure, forming a promoted sorbent. The promoter and the metal-oxygen-metal structures of the non-carbon base sorbent form metastable complexes, which are responsible for mercury capture via chemisorption. For example, metastable complexes may form between a halogen promoter and inorganic species on a non-carbon base sorbent, wherein inorganic species include sodium (Na), calcium (Ca), magnesium (Mg), aluminum (Al), iron (Fe) compounds, and combinations thereof. In some embodiments, the metastable complexes comprise a dihalogen group, such as Br-Cl, Br-Br, complexed with metal-oxygen-metal structures of the base sorbent. In some embodiments, the metastable complexes comprise a halogen atom, complexed with metal-oxygen-metal structures of the base sorbent. In some embodiments, the metastable complexes comprise a hydrohalogen group, complexed with metal-oxygen-metal structures of the base sorbent. In some embodiments, the metastable complexes comprise a Group V or Group VI halide, complexed with metal-oxygen-metal structures of the base sorbent. In some embodiments, the metal-oxygen-metal structures of the base sorbent are complexed with combinations of the functional groups disclosed herein. Without wishing to be limited by a theory, it is believed that mercury capture via chemisorption takes place through the action of mercury oxidation provided by these metastable complexes.

[0065] In embodiments, a carbon base sorbent with Lewis acid sites/groups comprising graphene sheets is activated by a promoter of this disclosure, forming a promoted sorbent. The promoter and the grapheme sheets of the carbon base sorbent form stable compounds, which are responsible for mercury capture via chemisorption. Without being limited by a theory, it is believed that mercury capture via chemisorption takes place through the action of mercury oxidation provided by these stable compounds. As one skilled in the art would appreciate, the versatility of chemistry associated with the base sorbent (carbon, non-carbon, or combination thereof) and the promoter of this disclosure enables versatile applications of the promoted sorbent system for mercury

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capture. This is especially advantageous because mercury content in flue gases varies from facility to facility, from operation to operation, and from day to day.

[0066] Examples of non-carbon base sorbents are amorphous forms of tectosilicates that comprise nanoscale cavities lined with Lewis basic oxygen associated with Group I alkali metals and Group II alkaline-earth metals. Such tectosilicates can be found in naturally occurring minerals, including, but not limited to, perlite and pumacite. Examples of non-carbon base sorbents also include amorphous forms of phyllosilicates. It is appreciated that other minerals may be used and also treated chemically and thermally to increase the activity of the base sorbent materials, such as phyllosilicates in the amorphous forms.

[0067] In some embodiments, bentonites are used as non-carbon base sorbents, including sodium bentonite and calcium bentonite. The use of other types of bentonites is contemplated as is known to one skilled in the art. The application of bentonite base sorbents is by introducing them into the flue gas at a location of the mercury capture system wherein the temperature of that location is below 800°C.

[0068] In some embodiments, the base sorbents are treated chemically and/or thermally to increase their activity. For example, perlite as a base sorbent may go through vacuum treatment and then thermal treatment so as to reduce the moisture contained therein, increase its activity, and potentially alter its morphology. Other treatment processes include chemical treatment, such acid treatment and alkaline treatment. These treatment methods may be combined to achieve desired effects as known to one skilled in the art. One of the desired effects is to increase the number of available Lewis acid sites/groups in the carbon base sorbent and/or the number of available Lewis basic sites/groups in the non-carbon base sorbent for subsequent activation via reaction with a promoter disclosed herein.

[0069] Referring now to Figure 3, a schematic flow diagram is provided of mercury control system 100 comprising preparation of promoted base sorbents and flue gas mercury reduction in accordance with preferred embodiments of the present invention. In the exemplary embodiment shown, there is provided a base sorbent reservoir 110, a halogen/halide promoter reservoir 120, a secondary component reservoir 130, and an alkali component reservoir 180, each of which with corresponding flow control device(s) 201, 202, 203, and 208/209, respectively. In conjunction with the alkali component reservoir 180, flow control devices 208 and 209 can be used independently, together, or not at all. Further, reservoirs 110, 120, 130 and 180 are optional and can be used in any combination, or not at all, whereby the otherwise stored components can be

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introduced into the system by other means either together or independently. Further, the alkali and secondary components may not be used at all within the system, if so desired.

[0070] As shown, reservoirs 110, 120, 130, and 180 connect through their respective flow control devices and via associated piping, to transport line 115. Alkali component reservoir 180 may also connect, through respective flow control devices and via associated piping, to transport line 118. A source of air, nitrogen, or other transport gas(es) is provided by gas source 170 to transport line 115 for the purpose of entraining materials discharged from reservoirs 110, 120, 130, and 180 and injecting such materials, via injection point 116, into contaminated flue gas stream 15. A source of air, nitrogen, or other transport gas(es) may be provided by gas source 171 to transport line 118 for the purpose of entraining materials discharged from reservoirs 180 and injecting such materials, via injection point 119, into flue gas stream 15. Gas sources 170 and 171 may be the same or different, as desired. Alternatively, transport gas(es) may be provided to both transport lines 115 and 118 by gas source 170 (connection from source 170 to line 118 not shown). Although gas sources 170 and 171 are shown in Figure 3 as compressors or blowers, any source of transport energy known in the art may be acceptable, as will be appreciated by those of skill in the art.

[0071] For clarity, single injection points 116 or 119 are shown in Figure 3, although one skilled in the art will understand that multiple injection points and/or locations are within the scope of the present invention. In the embodiment shown, transport line 15 comprises multiple lines allowing for multiple injections and separate and/or combined injections of base sorbent 110 and promoter 120 and/or 130. One mode of operation, by example, comprises providing base sorbent 110 in a common line which is promoted inline "in-flight" using promoter 120 and/or 130 and injected at point 116. Another mode of operation, by example, comprises transport and injection of a base sorbent 110 in a separate line to a point downstream of the injection of promoter 120 and/or 130 in a line at point 116 which is upstream of injection of base sorbent 110, resulting in in-flight preparation at a promoted sorbent within stream 15.

[0072] In the exemplary embodiment shown, an optional optical density measuring device(s) 204 is connected to transport line 115 and/or 118 to provide signals representative of the optical density inside transport line 115 and/or 118 as a function of time.

[0073] Downstream from injection point 116 and 119 is provided particulate separator 140. By way of illustration and not limitation, particulate separator 140 may comprise one or more fabric filters, one or more ESPs, or other particulate removal devices as are

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known in the art. It should be further noted that more than one particulate separator 140 may exist, sequentially or in parallel, and that injection point 116 and 119 may be at multiple locations upstream and/or downstream of 140 when parallel, sequential, or combinations thereof exist. Particulate separator 140 produces at least a predominantly gaseous ("clean") stream 142 and a stream 141 comprising separated solid materials. A sorbent/ash separator 150 separates stream 141 into a largely ash stream 152 and a largely sorbent stream 151. Stream 151 may then preferably be passed to an optional sorbent regenerator 160, which yields a regenerated sorbent stream 161 and a waste stream 162.

[0074] An optional continuous emission monitor (hereinafter "CEM") 205 for mercury is provided in exhaust gas stream 35 to provide electrical signals representative of the mercury concentration in exhaust stream 35 as a function of time. The optional mercury CEM 205 and flow controllers 201, 202, 203, 208, and 209 are electrically connected via optional lines 207 (or wirelessly) to an optional digital computer (or controller) 206, which receives and processes signals and preferably controls the preparation and injection of promoted carbon sorbent into contaminated flue gas stream 15.

[0075] In operation, as example, promoted sorbent and/or an optional alkali component is injected into contaminated flue gas stream 15. After contacting the injected material with the contaminated flue gas stream 15, the injected material reduces the mercury concentration, transforming contaminated flue gas into reduced mercury flue gas, 25. The injected material is removed from the flue gas 25 by separator 140, disposed of or further separated by optional separator 150, and disposed of or regenerated by an optional regenerator 160, respectively. The reduced mercury clean flue gas stream 142 is then monitored for mercury content by an optional CEM 205, which provides corresponding signals to an optional computer/controller 206. Logic and optimization signals from 206 then adjust flow controllers 201, 202, 203, 208, and 209 to maintain the mercury concentration in exhaust stream 35 within desired limits, according to control algorithms well known in the art. Flow controllers 201, 202, 203, 208, and 209 can also be adjusted manually or by some other automated means to maintain the mercury concentration in exhaust stream 35 within desired limits, according to control algorithms well known in the art.

[0076] Referring still to Figure 3, several embodiments are illustrated for preparation and injection of promoted sorbents and/or alkali components in accordance with the present invention. Stream 111 provides for introduction of base sorbent from reservoir 110, as metered by flow controller 201 manually or under the direction of computer 206.

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The halogen/halide may be combined and react with the base sorbent according to any of several provided methods. The halogen/halide may preferably be combined via line 121 directly into transport line 115, within which it contacts and reacts with the base sorbent prior to injection point 116 or downstream at point 116. This results in in-flight preparation of a promoted sorbent in accordance with the invention. Further, the halogen/halide may be combined via line 121b with the base sorbent prior to entering transport line 115. Still further, the halogen/halide may be contacted and react with the base sorbent by introduction via line 121c into reservoir 110. This option is preferably employed when, for example, reservoir 110 comprises an ebulliated or flueidized bed of base sorbent, through which halogen/halide flows in gaseous form or as a vapor. Of course, the halogen/halide may also preferably be contacted with the base sorbent in liquid form or in a solvent, as discussed previously, and solvent removal (not shown in Figure 3) may then be provided if necessary as mentioned with respect to embodiments discussed with reference to Figure 1.

[0077] Similarly, the optional secondary component may be contacted and react directly in transport line 115 via line 131, or optionally as described above with respect to the halogen/halide, via lines 131b and 131c.

[0078] Similarly, the optional alkali component from 180 may either be added in transport line 115 directly, or may be injected separately by transport line 118, combining downstream in flue gas 15 for synergistic effects with the base sorbent, promoted sorbent, or optional secondary components. Being able to vary on-site the amount of the optional alkali component relative to base sorbent, promoted sorbent, or optional secondary components is a key feature to overcome and optimize for site-specific operating and flue gas conditions.

[0079] In some preferred embodiments wherein contacting between components and reaction is performed in a liquid or solvent phase, stirring of such liquid and/or slurry mixtures may be provided. In other embodiments, the halogen/halide promoter and optional secondary component(s) may preferably be sprayed in solution form into or on the base sorbent. In some such embodiments, drying, filtering, centrifugation, settling, decantation, or other solvent removal methods as are known in the art may then be provided.

[0080] In embodiments wherein the halogen/halide promoter is in gaseous or vapor form, it may be diluted in air, nitrogen, or other gas as appropriate. The halide/halogen gas, for example, gaseous HBr or Br_2, may be passed through an ebulliated or flueidized

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bed of granular or fibrous base sorbent, with the promoted sorbent so produced removed from the top of the bed via gas entrainment for injection.

[0081] In some embodiments, the secondary component(s) may preferably comprise iodine or other halogens, hydrohalides, including without limitation HI, HBr, HCl, a Group V or Group VI element with a molecular halogen, such as SCl_2 and others. In some preferred embodiments, the promoted sorbent may comprise from about 1 to about 30 g of halogen/halide per 100 g of base sorbent. In some preferred embodiments, the promoted sorbent may comprise a secondary component in concentration of from about 1 to about 15 wt% of the concentration of the halogen/halide component.

[0082] In still other embodiments, the product-promoted sorbent may be applied to a substrate. In other embodiments, such prepared substrate(s) may be caused to contact a contaminated flue gas or gasification system product gas stream for mercury reduction purposes. Such substrates may be monolithic, rotating, or exposed to the gas stream in any number of ways known to those skilled in the art.

[0083] In some embodiments, a method is provided whereby a mercury stabilizing reagent is added to a promoted sorbent to produce a bifunctional sorbent. Such stabilizing reagent(s) may be sequentially added, either before or after the addition and reaction of the halogen/halide. In some preferred embodiments, the halogen/halide preferably comprises Br or HBr, and the mercury-stabilizing reagent may comprise S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof.

Halogens in Mercury Capture

[0084] Methodologies for using halogens for the treatment of flue gas have been problematic, owing to their reactivity with other gases and metals, resulting in corrosion and health issues. A "halogen" is defined as a member of the very active elements comprising Group VIIA (CAS nomenclature is used throughout; Group VIIA (CAS) corresponds to Group VIIB (IUPAC)) of the periodic table. In the molecular elemental form of the halogens, including F_2, Cl_2, Br_2, and I_2, the reaction with hot flue gas components leaves little to react with elemental mercury. The atomic elemental halogen form, which includes the fluorine, chlorine, bromine, and iodine atoms, is about a million times more reactive to mercury, but the concentration of the atomic forms is typically extremely low. In a large portion of electric utility coal combustion facilities, the concentrations are generally not sufficient to oxidize a significant amount of mercury.

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[0085] The term "halide" as used herein is defined as a compound formed from the reaction of a halogen with another element or radical. In general, halide compounds are much less reactive than the molecular halogens, having a low chemical potential. Halides are considered reduced forms that do not, alone, oxidize other compounds. In the conventional view, therefore, a halide salt-treated sorbent will not effectively oxidize elemental mercury and capture elemental mercury.

Halogen-Promoted Sorbent Characteristics

[0086] The promoted sorbent described here has a very high initial reactivity for oxidizing mercury and therefore, can be used in very small amounts to achieve very high capture efficiencies, thus lowering operation costs and lessening waste disposal problems. In addition, further disposal reductions are obtainable by regenerating and reusing the promoted sorbents produced using the inventive technology. The time interval required for the mercury and the promoted sorbents of the present invention to successfully interact in a flue gas duct, with subsequent collection of the mercury on the promoted sorbent and ash, is very short – less than seconds. Clearly, such collection times require the promoted sorbent to have both high capacity and high reactivity toward mercury. The promoted sorbent can be utilized in a very finely powdered form to minimize mass-transfer limitations. However, again, the reactivity should be very high to capture all of the mercury encountered by the fine particles. Additionally, use of these enhancement technologies allows capture to be effective for larger sorbent particles which also allows separation of the promoted sorbent from the ash to enable subsequent regeneration as well as ash utilization. One feature of this invention is the process to prepare a promoted sorbent containing a halide component formed on and/or within the base sorbent structure that provides a sorbent that is highly active on initial contact with the mercury-contaminated gas stream, which allows for very effective capture of the mercury.

[0087] The inventive sorbents chemically combine bromine species with Lewis acid/basic sites on the base sorbent. For example, x-ray photoelectron spectroscopy has established that the addition of bromine, chlorine, HBr, or HCl formed a chemical compound in the carbon structure. Thus the promoted sorbent produced from halogen and base sorbent does not represent a molecular halogen form, but rather a new chemically modified structure. This phenomenon may not occur with the less reactive iodine, where an I_2 molecular complex can exist on the carbon basal plane. In the case of bromine, modified cationic carbon has a high chemical potential for oxidation of

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mercury. Thus an entirely new model is presented for the reactivity of the bromine-treated carbon with mercury shown in Figure 2. The reactive carbon form can preferably be generated by the addition of bromine, hydrogen bromide, or combinations of bromine and other elements, as described herein. Halogen treatment resulted in higher-activity carbons because the halide anions (especially bromide and iodide) were effective in promoting oxidation by stabilizing the developing positive charge on the mercury in the transition state for oxidation. Based on this model, several innovative, inexpensive, activity-enhancing features have been developed.

Optional Second Component

[0088] It has been demonstrated that addition of an optional second component, in addition to the bromine, results in improved reactivity and capacity for the promoted sorbent, typically exceeding that of both the untreated base sorbent and the brominated carbon. The second compound comprises either a second halogen or a compound derived from a second halogen, such as HI. Thus in addition to having a reactive carbon form present, the second component generates a Lewis base with greater ability to stabilize the developing positive charge on the mercury. Thus the second component is an element with more polarized electrons (4p and 5p).

Optional Alkali Component

[0089] It has been demonstrated that addition of an optional alkali component with a base or promoted activated carbon results in improved mercury capture, typically exceeding that of both the untreated carbon and the promoted carbon. Test data indicate that flue gas contaminants, flue gas constituents (SO_2, NO_x, HCl, etc), operating temperature, mercury form, and mercury concentration may impact the effectiveness of the alkali addition. This suggests the need to be able to adjust and tailor the alkali-to-activated-carbon ratio on-site in order to overcome and optimize for a given set of site conditions.

[0090] The synergy that can be gained when coinjecting the two materials can be explained as follows. First, testing shows that binding sites on activated carbon (hereinafter "AC") can be consumed by chlorine species, sulfur species (i.e., sulfates), and other flue gas contaminants (arsenates, selenates, etc). The addition of optional alkali material will interact and react with these species/contaminants, thus minimizing their consumption of AC mercury binding sites. Second, testing also shows that standard AC will continue to oxidize mercury, even though the binding sites are fully

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consumed. This oxidized mercury can then react with alkali material and subsequently be captured by particulate control devices. Consequently, the addition of the optional alkali component acts to protect mercury-binding sites and capture oxidized mercury, thereby resulting in improved mercury reduction at lower cost. Alkali is generally much lower in cost (~ an order of magnitude less) than AC; thus more of it can be used, still resulting in overall lower costs.

In-Flight Sorbent Preparation

[0091] As stated previously, the halogen promoted sorbent can be readily produced in-flight. This is accomplished by, for example, contacting the vapors of any combination of halogens and, optionally, a second component, in-flight, with base sorbent particles. The particles may be dispersed in a stream of transport air (or other gas, such as the flue gas itself), which also conveys the halogen/halide-promoted sorbent particles to the flue gas duct, or other contaminated gas stream, from which mercury is to then be removed. There is no particular temperature requirement for this contact. This technology is obviously very simple to implement, and results in a great cost savings to facilities using this technology for mercury capture.

[0092] Referring to Figures 5A and 5B, process flow diagrams are shown as examples of the process for mercury removal from a mercury containing gas, such as a flue gas. In an embodiment shown at Figure 5A, gas inlet stream 501 passes through chamber 301 and enters air heater 302 as stream 502; then it exits air heater 302 as stream 503 and passes through particulate control device 303 and enters scrubber 304 as stream 504; finally it exits scrubber 304 as stream 505. As shown, chamber 301 is a boiler, however, one skilled in the art will appreciate that chamber 301 can also be the combustion chamber of a coal fired boiler, a stand alone combustion chamber or any other chamber in which mercury containing gas is either generated or passed through. Injection streams 401, 402, 403 and 404, in addition to stream 501, are multiple locations wherein promoter, base sorbent, or a combination of promoter and base sorbent may be introduced.

[0093] Figure 5B shows an embodiment similar to that of FIG. A, but with the position of the scrubber and particulate control device switched. Specifically, gas inlet stream 501' passes through chamber 301' and enters air heater 302' as stream 502'; then it exits air heater 302' as stream 503' and passes through scrubber 304' and enters particulate control device 303' as stream 504'; finally it exits particulate control device 303' as stream 505'. As shown, chamber 301 is a boiler, however, one skilled in the art will

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appreciate that chamber 301 can also be the combustion chamber of a coal fired boiler, a stand alone combustion chamber or any other chamber in which mercury containing gas is either generated or passed through. Injection stream locations 401', 402', 403' and 404', in addition to stream 501', represent multiple locations wherein promoter, base sorbent, or a combination of promoter and base sorbent may be introduced.

[0094] At each of the aforementioned injection stream locations 401, 401', 402, 402', 403, 403', 404 and 404', 501, 501' multiple injection points are contemplated so that promoter and base sorbent may be injected as a single injection stream or as separate injection streams, as further illustrated in the following exemplary embodiments.

[0095] In an embodiment, a promoter is introduced to chamber 301 at location 401. A base sorbent (carbon, non-carbon, or their combination) is introduced at location 402 upstream of air heater 302. In a further embodiment, a promoter is introduced to at location 402 upstream of air heater 302. A base sorbent (carbon, non-carbon, or their combination) is also introduced at location 402 either as a separate stream or as a mixed single stream with the promoter. In another embodiment, a promoter is introduced at locations 401 and 402. A base sorbent (carbon, non-carbon, or their combination) is introduced at location 402 either as a separate stream or as a mixed single stream with the promoter.

[0096] With the aid of this disclosure, one of ordinary skill in the art will be able to configure the process in many different fashions for mercury removal using the promoted sorbent. All these configurations are considered equivalents of the disclosed process and therefore are within the scope of the claimed invention.

Advantages of On-Site Preparation

[0097] In-flight preparation of the halogen/halide-promoted sorbent on location produces certain advantages. For example, the treatment system can be combined with the base sorbent injection system at the end-use site. With this technique, the halogen/halide is introduced to the base sorbent air (or other gas, including to the flue gas or other mercury containing gas) mixture in a transport line (or flue gas duct part of the base sorbent storage and injection system). This provides the following benefits over current conventional concepts for treating sorbents off-site:

- Capital equipment costs at a treatment facility are eliminated.
- Costs to operate the treatment facility are eliminated.
- There are no costs for transporting carbon and additive to a treatment facility.

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- The inventive process uses existing hardware and operation procedures.
- The inventive technology ensures that the sorbent is always fresh and, thus, more reactive.
- No new handling concerns are introduced.
- There are no costs for removing carbon from treatment system.
- The inventive process allows rapid on-site tailoring of additive–sorbent ratios in order to match the requirements of flue gas changes, such as may be needed when fuels are changed or loads are reduced, thus further optimizing the economics.
- The inventive technology reduces the amount of spent sorbents that are disposed.

[0098] With the foregoing and other features in view, there is provided, in accordance with the present invention, embodiments including a process for preparing and regenerating halogen/halide-promoted sorbents, whose activity for mercury capture is enhanced by the addition of halogen (e.g., bromine) to the base sorbent structure.

Sorbent Injection Location

[0099] Some of the preferred embodiments contemplate the use of a halogen-promoted sorbent in a powdered form that has been injected into a flue gas stream before or after ash particulates have been removed. Other embodiments of the inventive composition of the halogen-promoted sorbent comprise a powdered modified AC prepared by adding Br_2 or HBr plus a second optional component. Other embodiments allow the addition of the optional alkali component in conjunction with a base AC and/or with the use of a halogen-based sorbent and any other combinations of the sorbent technologies provided in this patent. Other embodiments allow for in-flight preparation of promoted sorbents by using and combining the promoters and base sorbents. Alternatively, embodiments include methods wherein the base sorbent is on a moving contactor consisting of particles or fibers containing one or more of the compositions listed above.

Sorbent Regeneration

[00100] Any of the above embodiments of the halogen/halide-promoted carbon and/or non-carbon sorbent can be easily regenerated; the poisoning contaminants from the flue gas are preferably removed, and an inexpensive promoting agent added, to restore mercury sorption activity. This process of promoting the activity of the carbon and/or non-carbon itself contrasts with the earlier, more expensive, conventional methods of

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adding a reagent (such as peroxide, gold, triiodide, etc.) to a base sorbent. The halogen/halide-promoted carbon sorbent of the present invention, treated with bromine and/or optional components, is noncorrosive. Detailed examples of sorbent regeneration techniques are described in copending, commonly owned PCT Patent Application No. PCT/US04/12828, titled "PROCESS FOR REGENERATING A SPENT SORBENT," which is hereby incorporated by reference in its entirety.

Sorbent Injection Control Schemes

[00101] Another advantage of the present invention relates to the use of a feedback system to more efficiently utilize certain aspects of the invention. Where possible and desirable, the mercury control technology of the present invention may preferably utilize continuous measurement of mercury emissions as feedback to assist in control of the sorbent injection rate. Tighter control on the sorbent and optional component(s) levels can be achieved in this way, which will ensure mercury removal requirements are met with minimal material requirements, thus minimizing the associated costs. In an embodiment, the mercury emissions are continuously measured downstream of the injection location, preferably in the exhaust gas at the stack.

EXAMPLES

[00102] To more clearly illustrate the present invention, an example is presented below. This example is intended to be illustrative, and no limitations to the present invention should be drawn or inferred from the example presented herein.

Example 1 – Non-carbon-Promoted Sorbent Tests at Full-Scale Plants

[00103] Full-scale commercial tests were conducted at several coal-burning facilities equipped with ESPs wherein three types of pre-cursers were injected upstream of an ESP at 325°F. In all, more than thirty tests were performed with variations in base sorbent material, base sorbent material injection rates and promoter injection rates. Promoters were halogen-based materials. Base sorbents were clay-based materials such as perlite and pumacite, which are vesicular forms of tectosilicates comprising silicates and aluminate tetrahedra with alkali metals in the interstitual spaces. Each base sorbent was injected upstream from the ESP.

[00104] The flue gas flow rate for the test was approximately 23 million $dsft^3/hr$ with a corresponding mercury flow rate of 0.0097 lbs Hg/hr. Initial mercury concentration in

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the flue gas prior to the introduction of the base sorbent or promoted sorbent was approximately 0.000417 lbs Hg/million dsft^3.

[00105] As shown in Table 1, each base sorbent was injected at three different test rates: about 100 lb/hr ("Low"), about 150 lb/hr ("Ave.") and about 200 lb/hr ("High"). At each test rate, the mercury removal rate was measured both with and without the use of a promoter to show the relative benefit of introducing the promoter into the flue gas stream. For the tests where promoter was injected into the flue gas stream, promoter was injected at about 20 lb/hr for the "Low" tests, at about 25 lb/hr for the "Ave." tests and from about 30 to about 50 lb/hr for the "High" tests. The percent mercury removal for each test was calculated based on measurements taken from the inlet flue gas and the outlet flue gas. The results as shown in Table 1 clearly show a significant improvement in mercury capture when a promoter and base sorbent are injected, as compared to the injection of a base sorbent alone.

Table 1. Test Data for Non Carbon-Promoted Sorbents

Base Sorbent Material	Base Sorbent Injection Rate (lb/hr)			Promoter Injection Rate (lb/hr)			Percent Removal of Mercury (%)		
	Low	Ave.	High	Low	Ave.	High	Low	Ave.	High
Perlite	100	150	200	0	0	0	37	40	43
	100	150	200	20	25	35	55	72	74
Punacite	100	150	200	0	0	0	25	25	25
	100	150	200	20	25	50	52	60	65
Clay-Based Mix of Silica and Alumina	100	150	200	0	0	0	50	52	54
	100	150	200	20	25	30	58	65	74

[00106] While the preferred embodiments of the invention have been shown and described, modifications thereof can be made by one skilled in the art without departing from the spirit and teachings of the invention. The embodiments described herein are exemplary only and are not intended to be limiting. Many variations and modifications of the invention disclosed herein are possible and are within the scope of the invention.

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Accordingly, the scope of protection is not limited by the description set out above, but is only limited by the claims which follow, that scope including all equivalents of the subject matter of the claims.

[00107] The examples provided in the disclosure are presented for illustration and explanation purposes only and are not intended to limit the claims or embodiment of this invention. While the preferred embodiments of the invention have been shown and described, modifications thereof can be made by one skilled in the art without departing from the spirit and teachings of the invention. Process criteria, equipment, and the like for any given implementation of the invention will be readily ascertainable to one of skill in the art based upon the disclosure herein. The embodiments described herein are exemplary only and are not intended to be limiting. Many variations and modifications of the invention disclosed herein are possible and are within the scope of the invention. Use of the term "optionally" with respect to any element of the invention is intended to mean that the subject element is required, or alternatively, is not required. Both alternatives are intended to be within the scope of the invention.

[00108] The discussion of a reference in the Background is not an admission that it is prior art to the present invention, especially any reference that may have a publication date after the priority date of this application. The disclosures of all patents, patent applications, and publications cited herein are hereby incorporated herein by reference in their entirety, to the extent that they provide exemplary, procedural, or other details supplementary to those set forth herein.

[00109] Although the invention is described herein as a promoted sorbent material and associated processes for its preparation and use, it is nevertheless not intended to be limited to the details described, 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.

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1. A method for separating mercury from a mercury containing gas comprising:

(a) providing a non-carbon sorbent material;

(b) providing a halogen or halide promoter;

(c) promoting at least a portion of the non-carbon sorbent material by chemically reacting the non-carbon sorbent material with the halogen or halide promoter to form a promoted halogenated sorbent;

(d) chemically reacting elemental mercury in the mercury containing gas with the promoted halogenated sorbent to form a mercury/sorbent chemical composition; and

(e) separating particulates from the mercury containing gas to form a cleaned gas, the particulates including ash and the first mercury/sorbent chemical composition.

2. The method of claim 1, further comprising the step of injecting the non-carbon sorbent material and the halogen or halide promoter into the mercury containing gas.

3. The method of claim 2, wherein said halogen or halide promoter and the non-carbon sorbent material are injected into the mercury containing gas at the same location.

4. The method of claim 2, wherein said halogen or halide promoter and the non-carbon sorbent material are injected into the mercury containing gas at separate locations.

5. The method of claim 4, wherein said halogen or halide promoter is injected into the mercury-containing gas upstream of the injection of said non-carbon sorbent material.

6. The method of claim 5, wherein said halogen or halide promoter is injected into a combustion chamber that produces a mercury-containing gas, and the non-carbon sorbent material is injected downstream of the combustion chamber.

7. The method of claim 6, wherein said halogen or halide promoter is additionally injected downstream of the combustion chamber.

8. The method of claim 6, wherein said combustion chamber is a boiler and the mercury-containing gas is a flue gas.

9. The method of claim 3, wherein said halogen or halide promoter and non-carbon sorbent material are injected downstream of a chamber that produces a mercury-containing gas.

10. The method of claim 9, wherein said chamber is a boiler and the mercury-containing gas is a flue gas.

11. The method of claim 2, wherein the rate at which said non-carbon sorbent material is injected or the rate at which said promoter is injected or combination thereof is adjusted according to a monitored mercury content in the cleaned gas so that the mercury content of the cleaned gas is maintained at substantially a desired level.

12. The method of claim 1, wherein said non-carbon sorbent material is selected from the group consisting of porous felsic materials, vesicular felsic materials, porous basaltic materials, vesicular basaltic materials, clay-based compounds, alkaline compounds, calcium hydroxide compounds, sodium acetate compounds, bicarbonate compounds, and combinations thereof.

13. The method of claim 12, wherein said non-carbon sorbent material is a material that reacts with oxidized mercury in the mercury-containing gas to form a second mercury/sorbent chemical composition.

14. The method of claim 1, wherein said non-carbon sorbent material comprises Lewis basic groups.

15. The method of claim 1, wherein said non-carbon sorbent material comprises amorphous forms of tectosilicates comprising nanoscale cavities lined with Lewis basic oxygen associated with alkaline-earth metals.

16. The method of claim 15, wherein said alkaline-earth metals comprise Group I and Group II alkaline-earth metals.

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17. The method of claim 1 wherein said non-carbon sorbent material comprises amorphous forms of phyllosilicates comprising nanoscale cavities lined with Lewis basic oxygen.

18. The method of claim 1, wherein said promoted sorbent comprises metastable complexes formed between said promoter and inorganic species on the non-carbon sorbent.

19. The method of claim 18 wherein said inorganic species is selected from the group consisting of sodium compounds, calcium compounds, magnesium compounds, aluminum compounds, iron compounds, and combinations thereof.

20. The method of claim 1, wherein said promoted sorbent comprises metastable complexes formed between said promoter and metal-oxygen-metal structures on the non-carbon sorbent.

21. The method of claim 20, wherein said promoter after being complexed with the metal-oxygen-metal structures is in the form selected from the group consisting of a dihalogen group, a halogen atom, a hydrohalogen group, a Group V halide, a Group VI halide, and combinations thereof.

22. The method of claim 1, wherein said promoted sorbent comprises activated Lewis basic groups or activated Lewis acid groups or combination thereof.

23. The method of claim 1, wherein said interaction between promoted sorbent and said mercury-containing gas stream comprises:

mercury diffusing from the gas phase onto said promoted sorbent surface; and

reacting with activated Lewis basic groups or activated Lewis acid groups or combination thereof to cause chemisorption on the sorbent surface.

24. The method of claim 1 further comprising pretreating said sorbent to increase the number of Lewis basic groups or Lewis acid groups or combination thereof on said sorbent.

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25. The method of claim 24, wherein pretreating said sorbent comprises chemical treatment, thermal treatment, vacuum treatment, and combinations thereof.

26. The method of claim 25, wherein said chemical treatment comprises acid treatment and alkaline treatment.

27. The method of claim 1 further comprising introducing an alkali component into the mercury-containing gas.

28. The method of claim 1, wherein a carbon sorbent is reacted with said promoter to produce a promoted carbon sorbent.

29. The method of claim 28, wherein said carbon sorbent or said promoter or combination thereof are introduced into the mercury-containing gas at one or more locations.

30. The method of claim 29, wherein the rate at which said carbon sorbent is introduced or the rate at which said promoter is introduced or combination thereof is adjusted according to a monitored mercury content in the cleaned gas so that the mercury content of the cleaned gas is maintained at substantially a desired level.

31. The method of claim 28, wherein said carbon sorbent comprises activated carbon.

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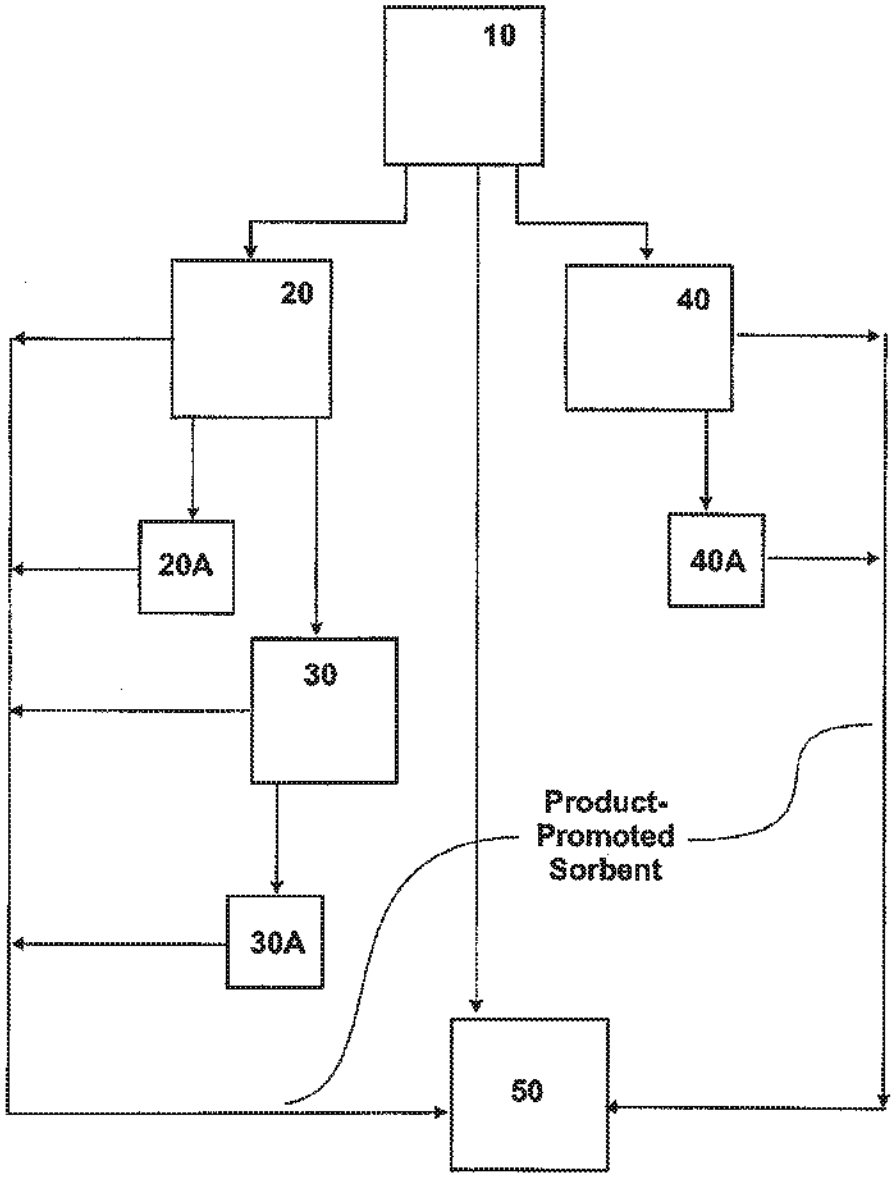


FIG. 1

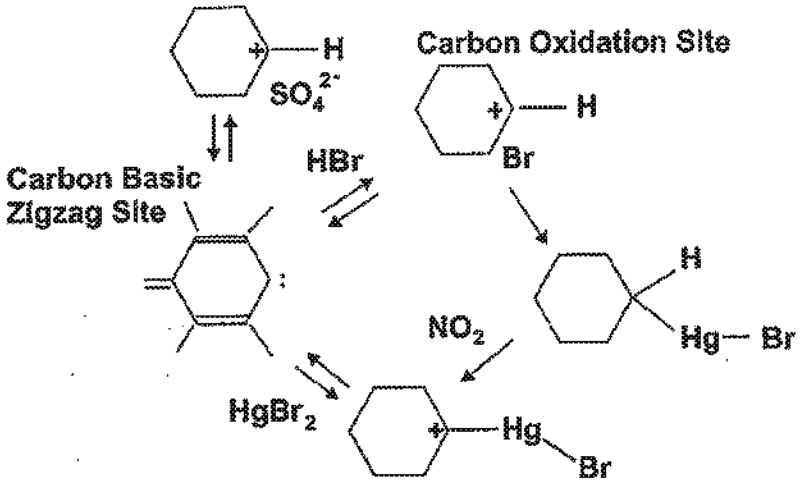

Carbon Oxidation Site
H
SO_4^{2-}
H
Br
HBr
Carbon Basic
Zigzag Site
H
Hg— Br
NO_2
$HgBr_2$
Hg
Br

FIG. 2

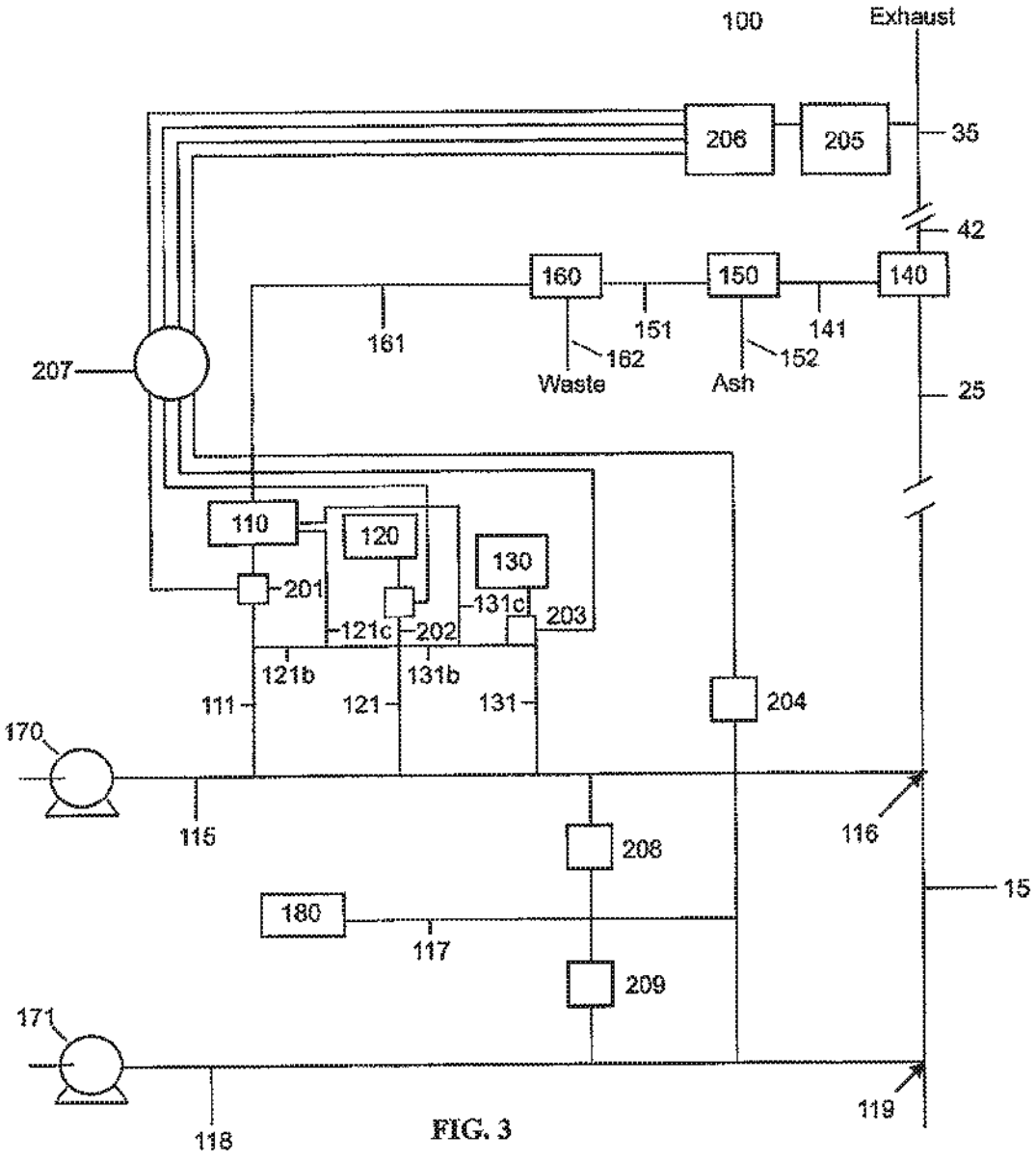

100
Exhaust
206
205
35
42
160
150
140
161
151
141
162
152
Waste
Ash
207
25
110
120
130
201
131c
203
121c
202
121b
131b
111
121
131
204
170
115
116
208
15
180
117
209
171
118
119

FIG. 3

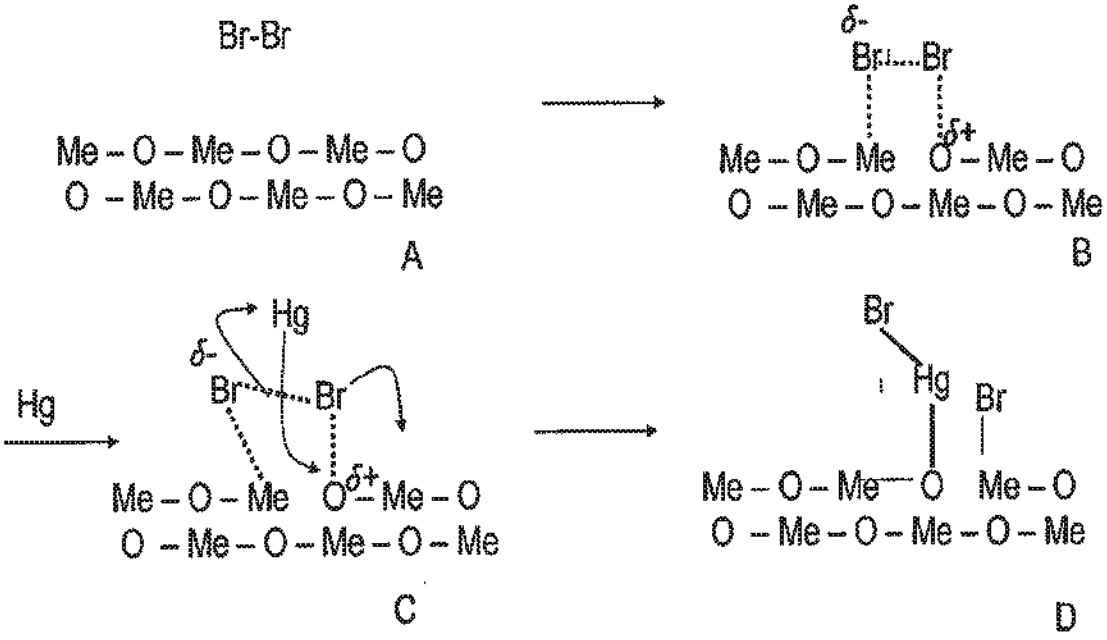

Br-Br
Me – O – Me – O – Me – O
O – Me – O – Me – O – Me
A
δ-
Br
Br
δ+
Me – O – Me
O – Me – O
Me – O – Me – O – Me
B
Hg
Hg
δ-
Br
Br
δ+
Me – O – Me
O – Me – O
Me – O
Me – O – Me – O – Me
C
Br
Hg
Br
Me – O – Me
O
Me – O
Me – O – Me – O – Me
D

FIG. 4

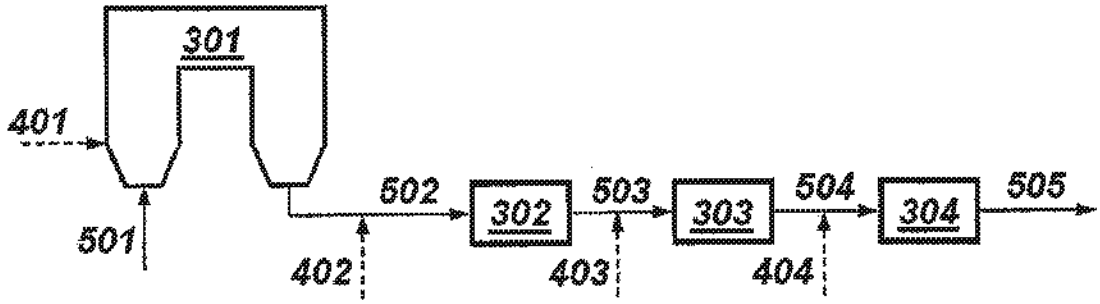

301
401
501
502
402
302
503
403
303
504
404
304
505

FIG. 5A

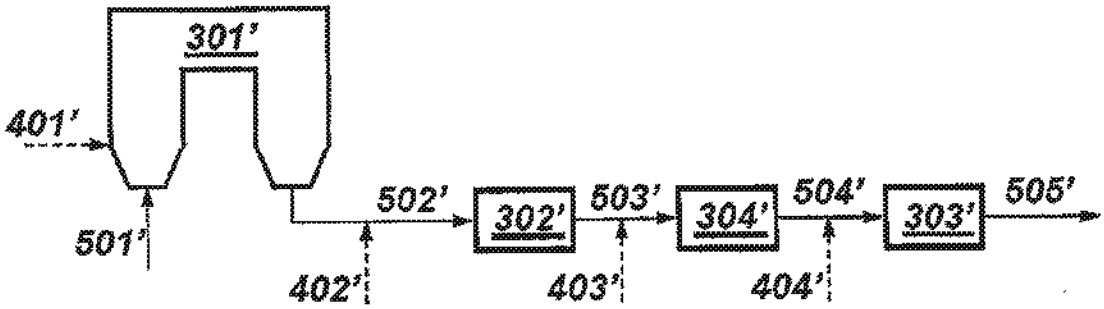

301'
401'
501'
502'
402'
302'
503'
403'
304'
504'
404'
303'
505'

FIG. 5B

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(54) Titre : SORBANTS POUR L'OXYDATION ET L'ELIMINATION DU MERCURE
(54) Title: SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

(57) Abrégé/Abstract:
A promoted carbon and/or non-carbon base sorbent are described that are highly effective for the removal of mercury from flue gas streams. The promoted sorbent comprises a carbon and/or non- carbon base sorbent that has reacted with and contains forms of halogen and halides. Optional components may be added to increase and/or preserve reactivity and mercury capacity. These may be added directly with the base sorbent, or in-flight within a gas stream (air, flue gas, etc.), to enhance base sorbent performance and/or mercury capture. Mercury removal efficiencies obtained exceed conventional methods. The promoted sorbent can be regenerated and reused. Base sorbent treatment and preparation methods are also described. New methods for in-flight preparation, introduction, and control of the active base sorbent into the mercury contaminated gas stream are described.

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(54) Title: SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

(57) Abstract: A promoted carbon and/or non-carbon base sorbent are described that are highly effective for the removal of mercury from flue gas streams. The promoted sorbent comprises a carbon and/or non-carbon base sorbent that has reacted with and contains forms of halogen and halides. Optional components may be added to increase and/or preserve reactivity and mercury capacity. These may be added directly with the base sorbent, or in-flight within a gas stream (air, flue gas, etc.), to enhance base sorbent performance and/or mercury capture. Mercury removal efficiencies obtained exceed conventional methods. The promoted sorbent can be regenerated and reused. Base sorbent treatment and preparation methods are also described. New methods for in-flight preparation, introduction, and control of the active base sorbent into the mercury contaminated gas stream are described.

SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

BACKGROUND OF THE INVENTION

Technical Field of the Invention

[0002] The present invention relates to methods and materials for the removal of pollutants from flue gas or product gas from a gasification system. In particular, mercury is removed from gas streams generated during the burning or gasification of fossil fuels by highly reactive regenerable sorbents.

Background of the Invention

[0003] The combustion and gasification of fossil fuel such as coal generates flue gas that contains mercury and other trace elements that originate from the fuel. The release of the mercury (and other pollutants) to the environment must be controlled by use of sorbents, scrubbers, filters, precipitators, and other removal technologies. Mercury is initially present in the elemental form during combustion and gasification. In downstream process sections, such as in the ducts and stack of a combustion system, some of the elemental mercury is oxidized. The amount that is oxidized depends on the amount of acid gases present in the flue gas and other factors. Amounts of mercury vary with the fuel, but concentrations of mercury in the stream of flue gas from coal combustion are typically less than 5 parts per billion (ppb). Large coal combustion facilities such as electric utilities may emit a pound of mercury, or more, a day. Mercury removal applications include, without limitation, flue gas from coal (or other fossil fuel) combustion, waste incineration, product gas from gasification, as well as offgases from mineral processing, metal refining, retorting, cement manufacturing, chloralkali plants, dental facilities, and crematories.

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Mercury Sorbent Technologies

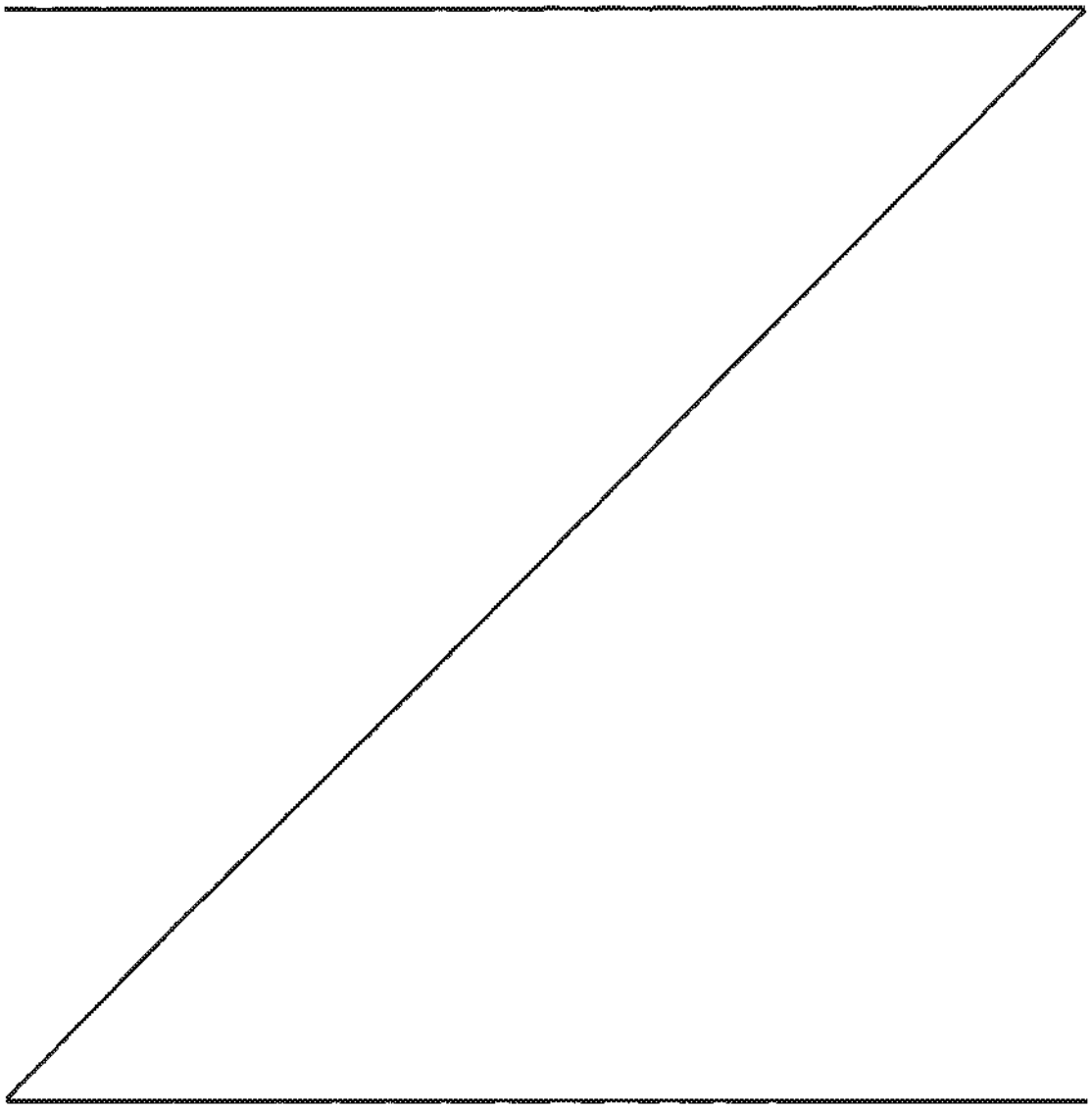
[0004] Several types of mercury control methods for flue gas have been investigated, including injection of fine sorbent particles into a flue gas duct and passing the flue gas through a sorbent bed. Fine-particle injection sorbents include activated carbon, metal oxide sorbent, sodium sulfide particles, and basic silicate or oxide sorbents. When particle injection is employed, the mercury captured on the sorbent particles is removed from the gas stream in a particulate control device such as a baghouse or electrostatic precipitator (ESP) and collected along with ash particulate. The sulfide and basic silicate and oxide particles are effective only for the oxidized mercury, and the metal oxide sorbents exhibit slower capture kinetics than the carbon particles. Additionally, injection of fine carbon particles into the flue gas stream has been only partially successful in removing mercury, especially elemental mercury, where effective removal of only about 60% is attained for some applications with a FF (fabric filter) to collect carbon and ash. Even lower removal rates have been observed when an ESP is used to collect the carbon because the contact time of the carbon with the gas is very short.

[0005] A major problem with existing carbon injection systems is that the sorbent is relatively unreactive toward mercury. Consequently, these sorbents must be used in large amounts, at high sorbent-to-mercury ratios, to effectively capture the mercury. These sorbents tend to be relatively expensive and cannot be easily separated from the ash for regeneration and reuse. The collection of carbon in the ash also creates solid waste disposal problems, and the spent sorbent may contaminate the collected ash, preventing its use in various applications.

[0006] One solution has been to add an oxidative sorbent comprising an aluminosilicate material impregnated with a very heavy dosage of one or more oxidative metal halides plus activated carbon. For example, refer to Varma et al. (published US application no. 2007/0140940). However, the amounts of metal salts required for Hg oxidation are generally relatively large and expensive. Also, several of the salts that can be used in such a process are highly toxic. Although the metal salts are present for oxidation in this process, activated carbon is essential for getting adsorption of the Hg.

As such, there is no synergistic role for the aluminosilicates as they appear to be only a support for the oxidizing salts.

[0007] Another approach has been the injection of aluminosilicate particulate such as bentonite, which contains neither oxidizing salts nor halogen complexes with a Lewis base site, and thus lacks the more powerful oxidizing capability of the said complexes as described in this application. For example, see U.S. Patent No. 7,413,719. Additionally, the injection of an aluminosilicate (kaolin or metakaolin) containing



calcium hypochlorite which thermally decomposes to form halogen is also known. For example, see U.S. Patent Application No. 20030103882. Thus these and similar impregnated aluminosilicate technologies require time in flight at appropriate high temperatures to heat the impregnated salt(s) to generate an oxidation site. This clearly represents a kinetic barrier to activation in contrast to the extremely fast complexing reaction of the Lewis acid on the surface of the appropriate Lewis base sorbent described in the present patent. The kinetic barrier is only for heating up the calcium hypochlorite to decompose it to Cl atoms or molecules. Halogen (bimolecular or atomic) would complex with carbon or noncarbon at any lower temperature to form reactive oxidation sites. Also, halide would require a very high temp or strong acid to form reactive halogen or halogen complex.

[0008] Yet another approach is the injection of bentonite plus a metal sulfide and a metal salt, none of which is oxidizing to elemental mercury and would require a slow thermal activation step. For example, see U.S. Patent Application No. 20070119300.

[0009] The injection of halogen or halogen precursors in a hot zone, followed by contact with an alkaline material in a wet or dry scrubber is another approach known in the art. With such an approach, elemental mercury is claimed to be oxidized by the halogen to Hg(II) which is collected by the alkaline material in the scrubber. For example, see U.S. Patent No 6,808,692 (Oehr), U.S. Patent No. 3,849,267 (Hilgen), U.S. Patent No. 5,435,980 (Felsvang), U.S. Patent No. 6,375,909 (Dangtran), U.S. Patent Application No. 20020114749 (Cole), U.S. Patent No. 6,638,485 (Iida), U.S. Patent Application No. 20030185718 (Sellakumar), U.S. Patent Application No. 20030147793 (Breen), and U.S. Patent No. 6,878,358 (Vosteen). However, even though it is known to inject halogen forms at some stage of the combustion process, such a process does not utilize a complexing method on a sorbent surface for conducting the oxidation and capture. Further, the alkaline material is rapidly surface-coated by the large concentrations of acid gases, lowering its capacity for adsorption of Hg(II). It is also recognized that the halogen forms initially introduced or generated are far more reactive to the large concentrations of SO_2 and moisture in the flue gas, and so gas-phase reactions of the halogens with Hg are hindered. In contrast, the present invention takes advantage of the Lewis acid complexes that rapidly form on the sorbent surface to effect the Hg oxidation, rather than rely on gas phase reactions for oxidation. Thus HCl, HBr, SO_2Br, and other gas-phase products all festoon the surface and promote the activity of the sorbent by forming complexes with the sorbent to form a promoted sorbent.

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[0010] Accordingly, there remains a need for more economical and effective mercury removal technology. This invention provides for cost-effective removal of poilutants, including mercury, using sorbent enhancement additives and/or highly reactive sorbents, with contact times of seconds (or less), and that may be regenerated and reused.

SUMMARY

[0011] The various embodiments of the present invention overcome the various aspects of the deficiencies of the prior art and provide new and economical methods for the removal of mercury from the gases produced in the utilization of fossil fuels.

[0011-a] An embodiment of the invention relates to a method for separating mercury from a mercury containing gas comprising:

(a) providing a carbon sorbent material and a non-carbon sorbent material, wherein the carbon sorbent material comprises activated carbon;

(b) providing a halogen or halide promoter;

(c) promoting at least a portion of the carbon sorbent material and the non-carbon sorbent material by chemically reacting the carbon sorbent material and the non-carbon sorbent material with the halogen or halide promoter to form a promoted halogenated carbon sorbent and a promoted halogenated non-carbon sorbent;

(d) chemically reacting elemental mercury in the mercury containing gas with the promoted halogenated carbon sorbent and the promoted halogenated non-carbon sorbent to form a mercury/sorbent chemical composition; and

(e) separating particulates from the mercury containing gas to form a cleaned gas, the particulates including ash and the mercury/sorbent chemical composition.

[0011-b] Another embodiment of the invention relates to the method defined hereinabove, further comprising the step of injecting the non-carbon sorbent material and the halogen or halide promoter into the mercury containing gas.

[0011-c] Another embodiment of the invention relates to the method defined hereinabove, wherein said halogen or halide promoter and the non-carbon sorbent material are injected into the mercury containing gas at the same location.

[0011-d] Another embodiment of the invention relates to the method defined hereinabove,

wherein said halogen or halide promoter and the non-carbon sorbent material are injected into the mercury containing gas at separate locations.

[0011-e] Another embodiment of the invention relates to the method defined hereinabove, wherein said halogen or halide promoter is injected into the mercury-containing gas upstream of the injection of said non-carbon sorbent material.

[0011-f] Another embodiment of the invention relates to the method defined hereinabove, wherein said halogen or halide promoter is injected into a combustion chamber that produces a mercury-containing gas, and the non-carbon sorbent material is injected downstream of the combustion chamber.

[0011-g] Another embodiment of the invention relates to the method defined hereinabove, wherein said halogen or halide promoter is additionally injected downstream of the combustion chamber.

[0011-h] Another embodiment of the invention relates to the method defined hereinabove, wherein said combustion chamber is a boiler and the mercury-containing gas is a flue gas.

[0011-i] Another embodiment of the invention relates to the method defined hereinabove, wherein said halogen or halide promoter and non-carbon sorbent material are injected downstream of a chamber that produces a mercury-containing gas.

[0011-j] Another embodiment of the invention relates to the method defined hereinabove, wherein said chamber is a boiler and the mercury-containing gas is a flue gas.

[0011-k] Another embodiment of the invention relates to the method defined hereinabove, wherein the rate at which said non-carbon sorbent material is injected or the rate at which said promoter is injected or combination thereof is adjusted according to a monitored mercury content in the cleaned gas so that the mercury content of the cleaned gas is maintained at a desired level.

[0011-l] Another embodiment of the invention relates to the method defined hereinabove, wherein said non-carbon sorbent material is selected from the group consisting of porous felsic materials, vesicular felsic materials, porous basaltic materials, vesicular basaltic materials, clay-based compounds, alkaline compounds, calcium hydroxide compounds, sodium acetate compounds, bicarbonate compounds, and combinations thereof.

[0011-m] Another embodiment of the invention relates to the method defined hereinabove, wherein said non-carbon sorbent material is a material that reacts with oxidized mercury in the mercury-containing gas to form a second mercury/sorbent chemical composition.

[0011-n] Another embodiment of the invention relates to the method defined hereinabove, wherein said non-carbon sorbent material comprises Lewis basic groups.

[0011-o] Another embodiment of the invention relates to the method defined hereinabove, wherein said non-carbon sorbent material comprises amorphous forms of tectosilicates comprising nanoscale cavities lined with Lewis basic oxygen associated with alkaline-earth metals.

[0011-p] Another embodiment of the invention relates to the method defined hereinabove, wherein said alkaline-earth metals comprise Group I and Group II alkaline-earth metals.

[0011-q] Another embodiment of the invention relates to the method defined hereinabove, wherein said non-carbon sorbent material comprises amorphous forms of phyllosilicates comprising nanoscale cavities lined with Lewis basic oxygen.

[0011-r] Another embodiment of the invention relates to the method defined hereinabove, wherein said promoted non-carbon sorbent comprises metastable complexes formed between said promoter and inorganic species on the non-carbon sorbent.

[0011-s] Another embodiment of the invention relates to the method defined hereinabove, wherein said inorganic species is selected from the group consisting of sodium compounds, calcium compounds, magnesium compounds, aluminum compounds, iron compounds, and combinations thereof.

[0011-t] Another embodiment of the invention relates to the method defined hereinabove, wherein said promoted non-carbon sorbent comprises metastable complexes formed between said promoter and metal-oxygen-metal structures on the non-carbon sorbent.

[0011-u] Another embodiment of the invention relates to the method defined hereinabove, wherein said promoter after being complexed with the metal-oxygen-metal structures is in the form selected from the group consisting of a dihalogen group, a halogen atom, a hydrohalogen group, a Group V halide, a Group VI halide, and combinations thereof.

[0011-v] Another embodiment of the invention relates to the method defined hereinabove,

wherein said promoted non-carbon sorbent comprises activated Lewis basic groups or activated Lewis acid groups or combination thereof.

[0011-w] Another embodiment of the invention relates to the method defined hereinabove, wherein said interaction between promoted non-carbon sorbent and said mercury-containing gas stream comprises:

mercury diffusing from the gas phase onto a surface of said non-carbon promoted sorbent; and

reacting with activated Lewis basic groups or activated Lewis acid groups or combination thereof to cause chemisorption on a sorbent surface.

[0011-x] Another embodiment of the invention relates to the method defined hereinabove, further comprising pretreating said non-carbon sorbent to increase the number of Lewis basic groups or Lewis acid groups or combination thereof on said sorbent.

[0011-y] Another embodiment of the invention relates to the method defined hereinabove, wherein pretreating said non-carbon sorbent comprises chemical treatment, thermal treatment, vacuum treatment, and combinations thereof.

[0011-z] Another embodiment of the invention relates to the method defined hereinabove, wherein said chemical treatment comprises acid treatment and alkaline treatment.

[0011-aa] Another embodiment of the invention relates to the method defined hereinabove, further comprising introducing an alkali component into the mercury-containing gas.

[0011-ab] Another embodiment of the invention relates to the method defined hereinabove, wherein said carbon sorbent or said promoter or combination thereof are introduced into the mercury-containing gas at one or more locations.

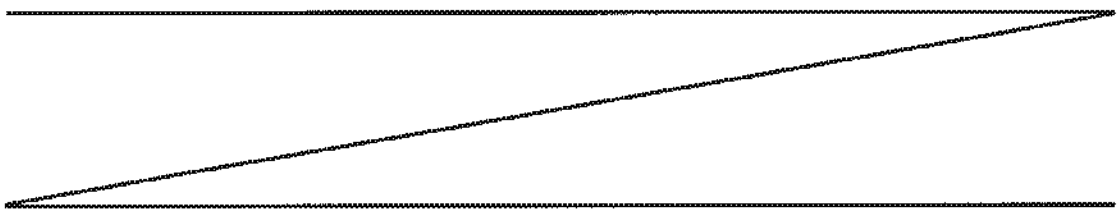
[0011-ac] Another embodiment of the invention relates to the method defined hereinabove, wherein the rate at which said carbon sorbent is introduced or the rate at which said promoter is introduced or combination thereof is adjusted according to a monitored mercury content in the cleaned gas so that the mercury content of the cleaned gas is maintained at a desired level.

[0011-ad] Another embodiment of the invention relates to the method defined hereinabove, where the promoted halogenated carbon sorbent and the promoted halogenated non-

carbon sorbent comprise 1 to 30 grams of the halogen or halide promoter per 100 grams of the carbon sorbent and the non-carbon sorbent.

[0012] A halogen/halide-promoted sorbent is described that is highly effective for the removal of mercury from flue gas streams. The sorbent comprises any activated carbon and/or non-carbon compound, such as porous or vesicular felsic or basaltic materials, clay-based compounds, alkaline compounds, calcium hydroxide compounds, sodium acetate compounds, and/or bicarbonate compounds, or a combination thereof. Optional secondary components and alkali may be added to further increase reactivity and mercury capacity. Mercury removal efficiencies obtained exceed or match conventional methods with added benefits such as reduced costs. Optionally, the promoted sorbent can be regenerated and reused. Sorbent treatment and/or preparation methods are also described. Methods for in-flight preparation, introduction, and control of the sorbent, promoter and promoted sorbent into the mercury contaminated gas stream are described.

[0013] When a promoted or a non-promoted base sorbent reacts with elemental or oxidized mercury, a mercury/sorbent chemical composition is formed and, in the case of elemental mercury reacting with the promoted base sorbent, the mercury is oxidized. As discussed in further detail in other portions of the specification, the base sorbent may be either a carbon or a non-carbon material or a combination thereof. Additionally, the mercury/sorbent chemical composition may be comprised of covalent bonds, ionic bonds and/or chemical complexes between the promoted or non-promoted base sorbent and the oxidized mercury. The Lewis basic groups on the non-promoted base sorbent, or the non-promoted portions of a promoted base sorbent, are available for reaction with the Lewis acid groups of already existing oxidized mercury in the mercury containing gas. Thus, mercury may be removed from the mercury containing gas stream through the formation of multiple and various mercury/sorbent chemical compositions even within the same process. For example, multiple sites on a sorbent particulate can form



multiple and various mercury/sorbent chemical compositions in the case where only a portion of the sites on the base sorbent particulate are promoted.

[0014] In some embodiments, a carbon and/or non-carbon promoted sorbent and/or a combination thereof is provided comprising a sorbent structure that has reacted with a promoter selected from the group consisting of halides, halogens, and combinations thereof, such that the reaction product is effective for the removal of mercury from a gas stream. The carbon sorbent comprises reactive Lewis acid groups/sites; the non-carbon sorbent comprises reactive Lewis basic groups/sites.

[0015] In an embodiment, a promoted carbon and/or non-carbon sorbent is provided wherein the base sorbent is selected from the group consisting of carbon, activated carbon, porous or vesicular felsic and basaltic materials, clay-based compounds, alkaline compounds, calcium hydroxide compounds, sodium acetate compounds, and/or bicarbonate compounds, or a combination thereof, with an average particle size similar to that of fly ash produced from a thermal process (combustion or gasification) or greater than that of fly ash produced such that it is physically separable therefrom, and combinations thereof, and the promoter is selected from the group consisting of molecular halogens, Group V (CAS nomenclature is used throughout) halides, Group VI halides, hydrohalides, and combinations thereof. In an embodiment, the promoted sorbent (carbon, non-carbon, or their combination) may have a mass mean particle diameter such that it can be substantially separated by physical means from entrained ash in the gas stream from which mercury is to be removed. In an embodiment, the base sorbent (carbon, non-carbon, or their combination) may have a mass mean particle diameter greater than about 40 micrometers.

[0016] In another embodiment, the promoted sorbent comprises from about 1 to about 30 grams of promoter per 100 grams of base sorbent. Another embodiment further comprises an optional secondary component comprising a halogen or a hydrohalide such that the reactivity and mercury capacity of the promoted sorbent are enhanced.

[0017] In another embodiment, the concentration of the optional secondary component on the finished sorbent is within the range of from about 1 to about 15 wt% of the concentration of the promoter on the finished sorbent.

[0018] In another embodiment, an optional alkali component may preferably be added to provide a synergistic effect through combination of this alkali with the base sorbent.

[0019] In another embodiment, the optional secondary component is selected from the group consisting of Group V halides, Group VI halides, HI, HBr, HCl, and combinations thereof. In another embodiment, the promoter is substantially in vapor

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form when combined with the base sorbent. In another embodiment, the promoter is combined with an organic solvent prior to reaction with the base sorbent. In another embodiment, the promoter and optional secondary component are combined with the base sorbentsubstantially simultaneously. Another embodiment further comprises adding a mercury-stabilizing reagent selected from the group consisting of S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof. Another embodiment further comprises adding an optional alkali component.

[0020] In an embodiment, a method is provided comprising providing a granular base sorbent and reacting the base sorbent with a promoter selected from the group consisting of halogens, halides, and combinations thereof, such that the reaction product comprises a promoted sorbent effective for removal of mercury from a gas stream. In a further embodiment, the reaction product comprises from about 1 to about 30 grams of promoter per 100 grams of base sorbent. In another embodiment the reaction product has an average particle-size distribution greater than the average size of entrained ash particles in the gas stream from which mercury is to be removed, such that the reaction product can be substantially removed from the entrained ash particles by physical means. In another embodiment, the reaction product has a mass mean particle diameter greater than about 40 micrometers.

[0021] In another embodiment, the promoter is selected from the group consisting of molecular halogens, hydrohalides, Group V halides, Group VI halides, and combinations thereof. In another embodiment, the promoter is in the gas phase when contacting the base sorbent (carbon, non-carbon, or their combination). In another embodiment, the promoter is in an organic solvent when contacting the base sorbent (carbon, non-carbon, or their combination).

[0022] In another embodiment, the promoter is selected from the group consisting of Br_2, a Group V bromide, a Group VI bromide, and combinations thereof.

[0023] In another embodiment, the method further comprises reacting the granular non-carbon with an optional secondary component comprising a halogen or a hydrohalide such that the reactivity and mercury capacity of the promoted sorbent are enhanced. In another embodiment, the promoter and optional secondary component are contacted simultaneously with the non-carbon base sorbent. In another embodiment, the method further comprises adding a mercury-stabilizing reagent selected from the group consisting of S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof. In an embodiment, a method is provided for control of mercury in a flue gas with substantially lower sorbent requirements. Through enhanced sorbent reactivity,

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mercury removal per gram of sorbent is increased, thereby decreasing the capital and operating costs by decreasing sorbent requirements.

[0024] In an embodiment, the promoted sorbent is introduced by direct injection into the flue gas stream. In another embodiment, the base sorbent is promoted within the flue gas stream

[0025] In an embodiment, a method is provided for reducing mercury in flue gas comprising providing a base sorbent, either by injection or in situ creation, into a mercury-containing flue gas stream, collecting greater than 70 wt% of the mercury in the flue gas on the promoted sorbent to produce a cleaned flue gas, and substantially recovering the promoted sorbent from the cleaned flue gas. In embodiments where less than 70 wt% mercury removal is desired, the required removal is attained using less base sorbent as would be required with standard base sorbent. In a further embodiment, the method further comprises monitoring the mercury content of the clean flue gas, regenerating the recovered promoted sorbent, and using the monitored mercury content of the cleaned flue gas to control the rate of base sorbent and promoter. In another embodiment the injected promoted sorbent is prepared in-flight by reacting a base sorbent (carbon, non-carbon, or their combination) and a promoter within a pneumatic transport line from which the reaction product is injected to the mercury-containing flue gas stream.

[0026] In another embodiment, the promoter is selected from the group consisting of molecular halogens, halides, and combinations thereof. In another embodiment, the promoter is reacted in the gas phase or as a vapor. In another embodiment, the promoter is added at from about 1 to about 30 grams per 100 grams of the base sorbent (carbon, non-carbon, or their combination).

[0027] In another embodiment, the injected promoted sorbent is prepared in-flight by reacting a base sorbent (carbon, non-carbon, or their combination), a promoter, and an optional secondary component to enhance the reactivity and capacity of the promoted sorbent within a pneumatic transport line from which the reaction product is injected to the mercury-containing flue gas stream.

[0028] In another embodiment, the optional secondary component is selected from the group consisting of iodine, hydrohalides, Group V halides, Group VI halides, and combinations thereof. In another embodiment, the optional secondary component is added at from about 1 to about 15 wt% of the promoter content. In another embodiment, the method further comprises adding to the promoted sorbent a mercury-stabilizing reagent selected from the group consisting of S, Se, H_2S, SO_2, H_2Se, SeO_2,

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CS_2, P_2S_5, and combinations thereof.

[0029] In an embodiment, the method further comprises coinjecting an optional alkaline material, including without limitation alkaline and alkaline-earth components, to improve the efficiency of mercury capture by capturing oxidized mercury and/or capturing gaseous components that might otherwise reduce promoted sorbent capacity. In another embodiment, the optional alkaline material may preferably comprise calcium oxide, sodium carbonate, and the like, as are known in the art.

[0030] In another embodiment, the method further comprises using the monitored mercury content of the cleaned flue gas to control the composition of the promoted sorbent. In another embodiment, the promoted sorbent is prepared in-flight by reacting a base sorbent (carbon, non-carbon, or their combination) and a promoter within the flue gas stream or in a transport line from which the reaction product is injected to the mercury-containing flue gas stream, wherein the promoter is selected from the group consisting of molecular halogens, halides, and combinations thereof, wherein the promoter is reacted in the gas phase or as a vapor, wherein the promoter is added at from about 1 to about 30 grams per 100 grams of the base sorbent (carbon, non-carbon, or their combination), wherein the rate at which the promoter is added and the rate of promoted sorbent injection are determined by a digital computer based, at least in part, on the monitored mercury content of the cleaned flue gas.

[0031] In an embodiment, a method for reducing the mercury content of a mercury and ash-containing gas stream is provided wherein particulate carbon and/or non-carbon promoted sorbent with a mass mean size greater than 40 μm is injected into the gas stream, mercury is removed from the gas by the promoted sorbent particles, the promoted sorbent particles are separated from the ash particles on the basis of size, and the promoted sorbent particles are reinjected to the gas stream. In another embodiment, the mercury-containing promoted sorbent particles are regenerated to remove some or substantially all of the mercury. In another embodiment, an alkaline component is coinjected into the gas stream. In another embodiment, the promoted sorbent may further comprise a promoter. The promoter may preferably comprise a halide, a halogen, or both.

[0032] In an embodiment, a method for reducing mercury in a mercury-containing gas to a desired level is disclosed. The method comprises reacting a carbon base sorbent with at least one promoter selected from the group consisting of molecular halogens, halides, and combinations thereof to produce a promoted carbon sorbent; allowing said promoted carbon sorbent to interact with a mercury-containing gas to capture mercury

in the mercury-containing gas on the promoted sorbent to produce a cleaned gas; and monitoring the mercury content of the cleaned gas. In some embodiments, the carbon base sorbent and the promoter are introduced into the mercury-containing gas at the same location or at separate locations. In some embodiments, the carbon base sorbent or promoter or combination thereof is introduced into the mercury-containing gas at one or more locations. In an embodiment, the rate at which the carbon base sorbent is introduced or the rate at which the promoter is introduced or combination thereof is adjusted according to the monitored mercury content of the cleaned gas so that the mercury content of the cleaned gas is maintained at substantially the desired level with minimal operating cost.

[0033] In a further emobodiment, the method comprises reacting a non-carbon base sorbent with at least one promoter selected from the group consisting of molecular halogens, halides, and combinations thereof to produce a promoted non-carbon sorbent; allowing said promoted non-carbon sorbent to interact with a mercury-containing gas to capture mercury in the mercury-containing gas on the promoted sorbent to produce a cleaned gas; and monitoring the mercury content of the cleaned gas. In some embodiments, the non-carbon base sorbent and the promoter are introduced into the mercury-containing gas at the same location or at separate locations. In some embodiments, the non-carbon base sorbent or promoter or combination thereof is introduced into the mercury-containing gas at one or more locations. In an embodiment, the rate at which the non-carbon base sorbent is introduced or the rate at which the promoter is introduced or combination thereof is adjusted according to the monitored mercury content of the cleaned gas so that the mercury content of the cleaned gas is maintained at substantially the desired level with minimal operating cost.

[0034] In a further embodiment, a method for reducing mercury in a mercury-containing gas to a desired level is presented. The method comprises reacting a base sorbent with at least one promoter selected from the group consisting of molecular halogens, halides, and combinations thereof to produce a promoted sorbent, wherein said base sorbent is selected from the group consisting of a non-carbon material, a carbon material, and combination thereof; allowing said promoted sorbent to interact with a mercury-containing gas to capture mercury in the mercury-containing gas on the promoted sorbent to produce a cleaned gas; and monitoring the mercury content of the cleaned gas.

[0035] In some embodiments, the base sorbent and the promoter are introduced into the mercury-containing gas at the same location or at separate locations. In some

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embodiments, the base sorbent or promoter or combination thereof is introduced into the mercury-containing gas at one or more locations. In some embodiments, introducing the base sorbent and the promoter comprises injecting the base sorbent and the promoter into the mercury-containing gas. In some embodiments, the promoter is introduced into the mercury-containing gas upstream of the introduction of the base sorbent. In some embodiments, the promoter is introduced upstream of a boiler or a combustion chamber. In an embodiment, the rate at which the base sorbent is introduced or the rate at which the promoter is introduced or combination thereof is adjusted according to the monitored mercury content of the cleaned gas so that the mercury content of the cleaned gas is maintained at substantially the desired level with minimal operating cost.

[0036] In an embodiment, the base sorbent for the promoted sorbent is selected from the group consisting of carbon, activated carbon, porous felsic materials, vesicular felsic materials, porous basaltic materials, vesicular basaltic materials, clay-based compounds, alkaline compounds, calcium hydroxide compounds, sodium acetate compounds, bicarbonate compounds, and combinations thereof. In embodiments, the non-carbon material comprises Lewis basic groups and the carbon material comprises Lewis acid groups. In some cases, the non-carbon material comprises amorphous forms of tectosilicates comprising nanoscale cavities lined with Lewis basic oxygen associated with alkaline-earth metals. The alkaline-earth metals comprise Group I and Group II alkaline-earth metals. In some other cases, the non-carbon material comprises amorphous forms of phyllosilicates comprising nanoscale cavities lined with Lewis basic oxygen.

[0037] In an embodiment, the promoted sorbent comprises metastable complexes formed between the promoter of this disclosure and inorganic species on the non-carbon base sorbent. In some embodiments, the inorganic species is selected from the group consisting of sodium compounds, calcium compounds, magnesium compounds, aluminum compounds, iron compounds, and combinations thereof. In an embodiment, the promoted sorbent comprises metastable complexes formed between the promoter of this disclosure and metal-oxygen-metal structures on the non-carbon base sorbent. In some embodiments, the promoter after being complexed with the metal-oxygen-metal structures is in the form selected from the group consisting of a dihalogen group, a halogen atom, a hydrohalogen group, a Group V halide, a Group VI halide, and combinations thereof. In an embodiment, the promoted sorbent comprises activated Lewis basic groups or activated Lewis acid groups or combination thereof. In some embodiments, the interaction between promoted sorbent and said mercury-containing

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gas stream comprises mercury diffusing from the gas phase onto said promoted sorbent surface; and reacting with the activated Lewis basic groups or activated Lewis acid groups or combination thereof to cause chemisorption on the promoted sorbent surface.

[0038] In an embodiment, the method for reducing mercury in a mercury-containing gas to a desired level further comprises pretreating the base sorbent to increase the number of Lewis basic groups or Lewis acid groups or combination thereof. In some cases, the pretreating methods comprise chemical treatment, thermal treatment, vacuum treatment, and combinations thereof. In some embodiments, chemical treatment comprises acid treatment and alkaline treatment, hi an embodiment, the method for reducing mercury in a mercury-containing gas to a desired level further comprises introducing an alkali component into the mercury-containing gas.

[0039] As will be described in more detail below, the present invention thus provides several advantages over previously known techniques, including significantly more effective and economical mercury sorbents for effluent gases, advantageously applicable to treating gas streams from combustion and gasification systems.

[0040] The foregoing has outlined rather broadly the features and technical advantages of the present invention in order that the detailed description of the invention that follows may be better understood. Additional features and advantages of the invention will be described hereinafter that form the subject of the claims of the invention. It should be appreciated by those skilled in the art that the conception and specific embodiments disclosed may be readily utilized as a basis for modifying or designing other structures for carrying out the same purposes of the present invention.

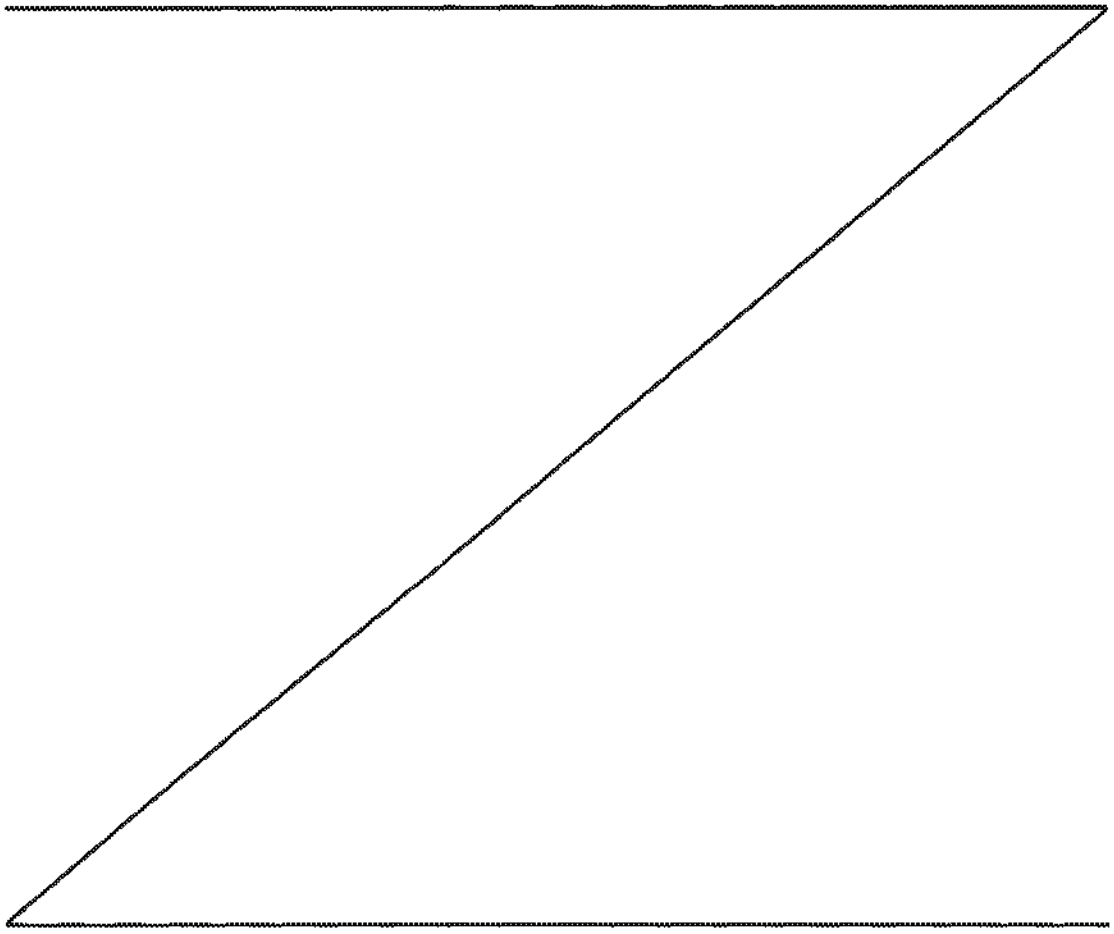
BRIEF DESCRIPTION OF THE DRAWINGS

[0041] For a more detailed description of the preferred embodiments of the present invention, reference will now be made to the accompanying drawings.

[0042] Figure 1 schematically illustrates methods for preparation of promoted carbon and/or non-carbon sorbents in accordance with the present invention.

[0043] Figure 2 illustrates a proposed mechanistic model of the chemical reactions resulting in the oxidation and capture of mercury.

[0044] Figure 3 schematically illustrates preparation of promoted carbon and/or non-carbon sorbents and processes for flue gas mercury reduction in flue gases and/or



product gases from a gasification system in accordance with the present invention, including in-flight preparation of promoted carbon and/or non-carbon sorbent.

[0045] Figure 4 illustrates a mechanism for promotion of metal oxide base sorbent via formation of a reactive halogen complex sorbent and subsequent capture of elemental mercury on the promoted sorbent.

[0046] Figure 5A schematically illustrates an exemplary process flow diagram for in-flight preparation of a promoted carbon and/or non-carbon sorbent.

[0047] Figure 5B schematically illustrates an exemplary process flow diagram for in-flight preparation of a promoted carbon and/or non-carbon sorbent.

DETAILED DESCRIPTION

[0048] Herein will be described in detail specific preferred embodiments of the present invention, with the understanding that the present disclosure is to be considered an exemplification of the principles of the invention and is not intended to limit the invention to that illustrated and described herein. The present invention is susceptible to preferred embodiments of different forms or order and should not be interpreted to be limited to the specifically expressed methods or compositions contained herein. In particular, various preferred embodiments of the present invention provide a number of different configurations and applications of the inventive method, compositions, and their uses.

[0049] The present invention provides a cost-effective way to capture pollutants by utilizing exceptionally reactive halogen/halide-promoted sorbents using a bromide (or other halogen/halide) treatment of the promoted sorbent, that capture mercury via mercury–sorbent surface reactions, at very short contact times of seconds or less. The reactivity of the promoted sorbent toward the pollutants (i.e., mercury) is greatly enhanced, and the sorption capacity can be regenerated; i.e., the promoted sorbent may be regenerated, recycled and/or reused.

[0050] The treated base sorbents (carbon, non-carbon, or their combination), treatment techniques, and optional additives discussed herein have applicability to mercury control from the product or effluent gas or gases from gasification systems, syngas generators, and other mercury-containing gas streams, in addition to the flue gas from combustion systems. Thus it should be understood that the terms combustion system and flue gas as used throughout this description may apply equally to gasification systems and syngas or fuel gas, as will be understood by those skilled in the art.

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[0051] Hereinafter the disclosure may at times discuss the use of carbon base sorbents in further details; however the use of non-carbon base sorbents and a combination of carbon and non-carbon base sorbents is also contemplated to at least the same degree as carbon base sorbents.

[0052] Referring now to Figure 1, there is shown a block flow diagram illustrating some preferred embodiments of the process of the present invention to prepare promoted sorbents useful for mercury capture in a mercury containing gas, such as a flue gas and/or product gas from gasification system streams. In a preferred embodiment illustrated by path 10-20, block 10 illustrates providing a base sorbent while block 20 illustrates adding a halogen or halide promoter that reacts with the base sorbent to produce a product-promoted sorbent. In embodiments where the halogen or halide is added, for example, as a vapor, no further steps may be necessary. In embodiments where the halogen or halide is added in, for example, a solvent, it may be desirable to employ solvent removal as illustrated by block 20A to produce a product-promoted sorbent suitable for injection.

[0053] Referring still to Figure 1, another preferred embodiment of the process of the present invention is illustrated by path 10-20-30, comprising providing a base sorbent as shown by block 10, adding a halogen or halide promoter that reacts with the base sorbent, illustrated at block 20, and adding a secondary component illustrated at block 30 that reacts with the result of block 20 to produce a product-promoted sorbent. In embodiments where both the halogen or halide promoter and the secondary component are added, for example, as a vapor, no further steps may be necessary. In embodiments where the halogen or halide promoter and/or secondary component are added in, for example, a solvent, it may be desirable to employ solvent removal as illustrated by block 30A to produce a product-promoted sorbent suitable for injection.

[0054] Referring still to Figure 1, another preferred embodiment of the process of the present invention is illustrated by path 10-40, comprising providing a base sorbent as illustrated at block 10 and adding a halogen or halide promoter and a secondary component to the base sorbent together, with which they react to produce a product-promoted sorbent as illustrated by block 40. As above, in embodiments where vapor additions are made to the base sorbent, no further steps may be desired. In embodiments where one or more components are added in solvent, a solvent removal step may be provided as illustrated by block 40A to produce a product-promoted sorbent suitable for injection.

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[0055] Referring still to Figure 1, another preferred embodiment of the process of the present invention is illustrated by path 10-50 in combination with path 20-50. In this embodiment, a base sorbent as illustrated by block 10 is introduced to the mercury containing gas as illustrated by block 50 while a halogen or halide promoter as illustrated by block 20 is introduced to the mercury containing gas stream. Thus the base sorbent and promoter react at block 50 to produce a product-promoted sorbent. In a similar manner, a secondary component as illustrated by block 30 may be added to the halogen or halide promoter as illustrated by block 20 and introduced into the mercury containing gas as illustrated by block 50. In embodiments where both the halogen or halide promoter and secondary component are added, for example, by vapor, no further steps may be taken. In embodiments where the halogen or halide and/or secondary component are added in, for example, a solvent, it may be desirable to employ solvent removal as illustrated by block 20A and/or block 30A.

[0056] Referring still to Figure 1, also illustrated are preferred embodiments in which, as illustrated by block 50, a mercury containing gas stream is treated with product-promoted carbon sorbent prepared as described above.

[0057] In some preferred embodiments, the carbon base sorbent provided may preferably be any of several types, as understood by those skilled in the art. For example, the carbon base sorbent may include powdered activated carbon, granular activated carbon, carbon black, unburned carbon, carbon fiber, carbon honeycomb or plate structure, aerogel carbon film, pyrolysis char, regenerated activated carbon from product-promoted carbon sorbent, or other types as known in the art.

[0058] In some preferred embodiments, the carbon base sorbent provided may preferably have a mass mean particle size greater than the fly ash in a mercury containing gas, such as a flue gas stream, to be treated.

[0059] In some preferred embodiments, the carbon base sorbent provided may preferably have a mass mean particle diameter preferably greater than 40 micrometers, more preferably greater than 60 micrometers, or a particle-size distribution greater than that of the fly ash or entrained ash in a flue gas stream, or other mercury containing gas, to be treated, such that the activated carbon and ash can be separated by physical means.

[0060] In some preferred embodiments, the halogen or halide promoter that is added to, and reacts with, the carbon base sorbent may preferably comprise, by way of illustration and not limitation, a molecular halogen in vapor or gaseous form, a molecular halogen in an organic solvent, a Group V or Group VI halide, such as PBr_3 or SCl_2, respectively, in vapor, liquid, or solution form (though not in an aqueous solvent).

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[0061] Embodiments are also provided in which the organic solvent may preferably comprise a chlorinated hydrocarbon, such as dichloromethane, a hydrocarbon solvent, including for example, petroleum ether, ligroin, pentane, hexane, toluene, and benzene, carbon disulfide, a waste solvent, an ether, a recycled solvent, a supercritical solvent, such as supercritical CO_2, water (though not in the case of a Group V or Group VI halide), and others as will be apparent to those of skill in the art.

[0062] Referring now to Figure 4, a theory is illustrated developed from scientific evidence to explain the nature of the promoting compounds. For example, as illustrated in Figure 4, a molecular bromine molecule forms a complex with the surface of the base sorbent comprising a high surface area form of a metal oxide. Complexing with bromine can occur at surface and defect sites on the surface of the glassy amorphous tectosilicates by association with the Lewis basic oxygen lining the interstitial cavities and the alkali actions on the surface. Molecular hydrogen bromide or an electrophilic or Lewis acid bromine compound react to form a similar structure. The precedence for this promotion of a metal oxide surface complex with an acidic species is described in a paper by Stark and Klabunde (Klabunde, K.J. Chem. Mater. 1996, 8, 1913-1918) who showed addition of acids, HCl, SO_3, and NO, to a MgO surface to form a surface complex. In US Patent 6,517,423 to Koper et al. the described surface complexes were active for destroying biological agents and toxins. As shown in Figure 4, addition of halogens to a metal oxide surface (A) can form a complex that could be described as a bromide-oxybromide species (B), in which electrophilic reactivity is maintained, owing to the formation of positive charges on the surface. The electrophilic complex formed on the metal oxide base sorbent comprises an active site for oxidation of elemental mercury. Addition of mercury to the complex results in formation of a mercury oxygen bond and, simultaneously, a mercury bromine bond as shown in complex C in Figure 4. Thus the final structure is a stable oxidized Hg form (D) described as a metaloxymercuric bromide.

[0063] In summary, it has now been found that the formation of the new bromide complex with the metal oxide surface increases the surface reactivity toward mercury and other pollutants. Additionally, the resulting bromide compound is uniquely suited to facilitate oxidation of the mercury. The effectiveness of the oxidation apparently results from the promotion effect of the halide, exerted on the developing positive charge on the mercury during the oxidation, known in the chemical art as a specific catalytic effect. Thus as the mercury electrons are drawn toward the positive surface oxygen, the halide anion electrons are pushing in from the other side, stabilizing the

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positive charge developing on the mercury and lowering the energy requirement for the oxidation process. Bromide is especially reactive, owing to the highly polarizable electrons in the outer 4p orbitals of the ion. Thus adding HBr or Br_2 to the appropriate metal oxide forms a similar surface bromide complex, in which the positive oxygen oxidizes the mercury with the assistance of the bromide ion.

[0064] In embodiments, a non-carbon base sorbent with Lewis basic sites/groups comprising metal-oxygen-metal structures is activated by a promoter of this disclosure, forming a promoted sorbent. The promoter and the metal-oxygen-metal structures of the non-carbon base sorbent form metastable complexes, which are responsible for mercury capture via chemisorption. For example, metastable complexes may form between a halogen promoter and inorganic species on a non-carbon base sorbent, wherein inorganic species include sodium (Na), calcium (Ca), magnesium (Mg), aluminum (Al), iron (Fe) compounds, and combinations thereof. In some embodiments, the metastable complexes comprise a dihalogen group, such as Br-Cl, Br-Br, complexed with metal-oxygen-metal structures of the base sorbent. In some embodiments, the metastable complexes comprise a halogen atom, complexed with metal-oxygen-metal structures of the base sorbent. In some embodiments, the metastable complexes comprise a hydrohalogen group, complexed with metal-oxygen-metal structures of the base sorbent. In some embodiments, the metastable complexes comprise a Group V or Group VI halide, complexed with metal-oxygen-metal structures of the base sorbent. In some embodiments, the metal-oxygen-metal structures of the base sorbent are complexed with combinations of the functional groups disclosed herein. Without wishing to be limited by a theory, it is believed that mercury capture via chemisorption takes place through the action of mercury oxidation provided by these metastable complexes.

[0065] In embodiments, a carbon base sorbent with Lewis acid sites/groups comprising graphene sheets is activated by a promoter of this disclosure, forming a promoted sorbent. The promoter and the grapheme sheets of the carbon base sorbent form stable compounds, which are responsible for mercury capture via chemisorption. Without being limited by a theory, it is believed that mercury capture via chemisorption takes place through the action of mercury oxidation provided by these stable compounds. As one skilled in the art would appreciate, the versatility of chemistry associated with the base sorbent (carbon, non-carbon, or combination thereof) and the promoter of this disclosure enables versatile applications of the promoted sorbent system for mercury

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capture. This is especially advantageous because mercury content in flue gases varies from facility to facility, from operation to operation, and from day to day.

[0066] Examples of non-carbon base sorbents are amorphous forms of tectosilicates that comprise nanoscale cavities lined with Lewis basic oxygen associated with Group I alkali metals and Group II alkaline-earth metals. Such tectosilicates can be found in naturally occurring minerals, including, but not limited to, perlite and pumacite. Examples of non-carbon base sorbents also include amorphous forms of phyllosilicates. It is appreciated that other minerals may be used and also treated chemically and thermally to increase the activity of the base sorbent materials, such as phyllosilicates in the amorphous forms.

[0067] In some embodiments, bentonites are used as non-carbon base sorbents, including sodium bentonite and calcium bentonite. The use of other types of bentonites is contemplated as is known to one skilled in the art. The application of bentonite base sorbents is by introducing them into the flue gas at a location of the mercury capture system wherein the temperature of that location is below 800°C.

[0068] In some embodiments, the base sorbents are treated chemically and/or thermally to increase their activity. For example, perlite as a base sorbent may go through vacuum treatment and then thermal treatment so as to reduce the moisture contained therein, increase its activity, and potentially alter its morphology. Other treatment processes include chemical treatment, such acid treatment and alkaline treatment. These treatment methods may be combined to achieve desired effects as known to one skilled in the art. One of the desired effects is to increase the number of available Lewis acid sites/groups in the carbon base sorbent and/or the number of available Lewis basic sites/groups in the non-carbon base sorbent for subsequent activation via reaction with a promoter disclosed herein.

[0069] Referring now to Figure 3, a schematic flow diagram is provided of mercury control system 100 comprising preparation of promoted base sorbents and flue gas mercury reduction in accordance with preferred embodiments of the present invention. In the exemplary embodiment shown, there is provided a base sorbent reservoir 110, a halogen/halide promoter reservoir 120, a secondary component reservoir 130, and an alkali component reservoir 180, each of which with corresponding flow control device(s) 201, 202, 203, and 208/209, respectively. In conjunction with the alkali component reservoir 180, flow control devices 208 and 209 can be used independently, together, or not at all. Further, reservoirs 110, 120, 130 and 180 are optional and can be used in any combination, or not at all, whereby the otherwise stored components can be

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introduced into the system by other means either together or independently. Further, the alkali and secondary components may not be used at all within the system, if so desired.

[0070] As shown, reservoirs 110, 120, 130, and 180 connect through their respective flow control devices and via associated piping, to transport line 115. Alkali component reservoir 180 may also connect, through respective flow control devices and via associated piping, to transport line 118. A source of air, nitrogen, or other transport gas(es) is provided by gas source 170 to transport line 115 for the purpose of entraining materials discharged from reservoirs 110, 120, 130, and 180 and injecting such materials, via injection point 116, into contaminated flue gas stream 15. A source of air, nitrogen, or other transport gas(es) may be provided by gas source 171 to transport line 118 for the purpose of entraining materials discharged from reservoirs 180 and injecting such materials, via injection point 119, into flue gas stream 15. Gas sources 170 and 171 may be the same or different, as desired. Alternatively, transport gas(es) may be provided to both transport lines 115 and 118 by gas source 170 (connection from source 170 to line 118 not shown). Although gas sources 170 and 171 are shown in Figure 3 as compressors or blowers, any source of transport energy known in the art may be acceptable, as will be appreciated by those of skill in the art.

[0071] For clarity, single injection points 116 or 119 are shown in Figure 3, although one skilled in the art will understand that multiple injection points and/or locations are within the scope of the present invention. In the embodiment shown, transport line 15 comprises multiple lines allowing for multiple injections and separate and/or combined injections of base sorbent 110 and promoter 120 and/or 130. One mode of operation, by example, comprises providing base sorbent 110 in a common line which is promoted inline "in-flight" using promoter 120 and/or 130 and injected at point 116. Another mode of operation, by example, comprises transport and injection of a base sorbent 110 in a separate line to a point downstream of the injection of promoter 120 and/or 130 in a line at point 116 which is upstream of injection of base sorbent 110, resulting in in-flight preparation at a promoted sorbent within stream 15.

[0072] In the exemplary embodiment shown, an optional optical density measuring device(s) 204 is connected to transport line 115 and/or 118 to provide signals representative of the optical density inside transport line 115 and/or 118 as a function of time.

[0073] Downstream from injection point 116 and 119 is provided particulate separator 140. By way of illustration and not limitation, particulate separator 140 may comprise one or more fabric filters, one or more ESPs, or other particulate removal devices as are

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known in the art. It should be further noted that more than one particulate separator 140 may exist, sequentially or in parallel, and that injection point 116 and 119 may be at multiple locations upstream and/or downstream of 140 when parallel, sequential, or combinations thereof exist. Particulate separator 140 produces at least a predominantly gaseous ("clean") stream 142 and a stream 141 comprising separated solid materials. A sorbent/ash separator 150 separates stream 141 into a largely ash stream 152 and a largely sorbent stream 151. Stream 151 may then preferably be passed to an optional sorbent regenerator 160, which yields a regenerated sorbent stream 161 and a waste stream 162.

[0074] An optional continuous emission monitor (hereinafter "CEM") 205 for mercury is provided in exhaust gas stream 35 to provide electrical signals representative of the mercury concentration in exhaust stream 35 as a function of time. The optional mercury CEM 205 and flow controllers 201, 202, 203, 208, and 209 are electrically connected via optional lines 207 (or wirelessly) to an optional digital computer (or controller) 206, which receives and processes signals and preferably controls the preparation and injection of promoted carbon sorbent into contaminated flue gas stream 15.

[0075] In operation, as example, promoted sorbent and/or an optional alkali component is injected into contaminated flue gas stream 15. After contacting the injected material with the contaminated flue gas stream 15, the injected material reduces the mercury concentration, transforming contaminated flue gas into reduced mercury flue gas, 25. The injected material is removed from the flue gas 25 by separator 140, disposed of or further separated by optional separator 150, and disposed of or regenerated by an optional regenerator 160, respectively. The reduced mercury clean flue gas stream 142 is then monitored for mercury content by an optional CEM 205, which provides corresponding signals to an optional computer/controller 206. Logic and optimization signals from 206 then adjust flow controllers 201, 202, 203, 208, and 209 to maintain the mercury concentration in exhaust stream 35 within desired limits, according to control algorithms well known in the art. Flow controllers 201, 202, 203, 208, and 209 can also be adjusted manually or by some other automated means to maintain the mercury concentration in exhaust stream 35 within desired limits, according to control algorithms well known in the art.

[0076] Referring still to Figure 3, several embodiments are illustrated for preparation and injection of promoted sorbents and/or alkali components in accordance with the present invention. Stream 111 provides for introduction of base sorbent from reservoir 110, as metered by flow controller 201 manually or under the direction of computer 206.

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The halogen/halide may be combined and react with the base sorbent according to any of several provided methods. The halogen/halide may preferably be combined via line 121 directly into transport line 115, within which it contacts and reacts with the base sorbent prior to injection point 116 or downstream at point 116. This results in in-flight preparation of a promoted sorbent in accordance with the invention. Further, the halogen/halide may be combined via line 121b with the base sorbent prior to entering transport line 115. Still further, the halogen/halide may be contacted and react with the base sorbent by introduction via line 121c into reservoir 110. This option is preferably employed when, for example, reservoir 110 comprises an ebulliated or flueidized bed of base sorbent, through which halogen/halide flows in gaseous form or as a vapor. Of course, the halogen/halide may also preferably be contacted with the base sorbent in liquid form or in a solvent, as discussed previously, and solvent removal (not shown in Figure 3) may then be provided if necessary as mentioned with respect to embodiments discussed with reference to Figure 1.

[0077] Similarly, the optional secondary component may be contacted and react directly in transport line 115 via line 131, or optionally as described above with respect to the halogen/halide, via lines 131b and 131c.

[0078] Similarly, the optional alkali component from 180 may either be added in transport line 115 directly, or may be injected separately by transport line 118, combining downstream in flue gas 15 for synergistic effects with the base sorbent, promoted sorbent, or optional secondary components. Being able to vary on-site the amount of the optional alkali component relative to base sorbent, promoted sorbent, or optional secondary components is a key feature to overcome and optimize for site-specific operating and flue gas conditions.

[0079] In some preferred embodiments wherein contacting between components and reaction is performed in a liquid or solvent phase, stirring of such liquid and/or slurry mixtures may be provided. In other embodiments, the halogen/halide promoter and optional secondary component(s) may preferably be sprayed in solution form into or on the base sorbent. In some such embodiments, drying, filtering, centrifugation, settling, decantation, or other solvent removal methods as are known in the art may then be provided.

[0080] In embodiments wherein the halogen/halide promoter is in gaseous or vapor form, it may be diluted in air, nitrogen, or other gas as appropriate. The halide/halogen gas, for example, gaseous HBr or Br_2, may be passed through an ebulliated or flueidized

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bed of granular or fibrous base sorbent, with the promoted sorbent so produced removed from the top of the bed via gas entrainment for injection.

[0081] In some embodiments, the secondary component(s) may preferably comprise iodine or other halogens, hydrohalides, including without limitation HI, HBr, HCl, a Group V or Group VI element with a molecular halogen, such as SCl_2 and others. In some preferred embodiments, the promoted sorbent may comprise from about 1 to about 30 g of halogen/halide per 100 g of base sorbent. In some preferred embodiments, the promoted sorbent may comprise a secondary component in concentration of from about 1 to about 15 wt% of the concentration of the halogen/halide component.

[0082] In still other embodiments, the product-promoted sorbent may be applied to a substrate. In other embodiments, such prepared substrate(s) may be caused to contact a contaminated flue gas or gasification system product gas stream for mercury reduction purposes. Such substrates may be monolithic, rotating, or exposed to the gas stream in any number of ways known to those skilled in the art.

[0083] In some embodiments, a method is provided whereby a mercury stabilizing reagent is added to a promoted sorbent to produce a bifunctional sorbent. Such stabilizing reagent(s) may be sequentially added, either before or after the addition and reaction of the halogen/halide. In some preferred embodiments, the halogen/halide preferably comprises Br or HBr, and the mercury-stabilizing reagent may comprise S, Se, H_2S, SO_2, H_2Se, SeO_2, CS_2, P_2S_5, and combinations thereof.

Halogens in Mercury Capture

[0084] Methodologies for using halogens for the treatment of flue gas have been problematic, owing to their reactivity with other gases and metals, resulting in corrosion and health issues. A "halogen" is defined as a member of the very active elements comprising Group VIIA (CAS nomenclature is used throughout; Group VIIA (CAS) corresponds to Group VIIB (IUPAC)) of the periodic table. In the molecular elemental form of the halogens, including F_2, Cl_2, Br_2, and I_2, the reaction with hot flue gas components leaves little to react with elemental mercury. The atomic elemental halogen form, which includes the fluorine, chlorine, bromine, and iodine atoms, is about a million times more reactive to mercury, but the concentration of the atomic forms is typically extremely low. In a large portion of electric utility coal combustion facilities, the concentrations are generally not sufficient to oxidize a significant amount of mercury.

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[0085] The term "halide" as used herein is defined as a compound formed from the reaction of a halogen with another element or radical. In general, halide compounds are much less reactive than the molecular halogens, having a low chemical potential. Halides are considered reduced forms that do not, alone, oxidize other compounds. In the conventional view, therefore, a halide salt-treated sorbent will not effectively oxidize elemental mercury and capture elemental mercury.

Halogen-Promoted Sorbent Characteristics

[0086] The promoted sorbent described here has a very high initial reactivity for oxidizing mercury and therefore, can be used in very small amounts to achieve very high capture efficiencies, thus lowering operation costs and lessening waste disposal problems. In addition, further disposal reductions are obtainable by regenerating and reusing the promoted sorbents produced using the inventive technology. The time interval required for the mercury and the promoted sorbents of the present invention to successfully interact in a flue gas duct, with subsequent collection of the mercury on the promoted sorbent and ash, is very short – less than seconds. Clearly, such collection times require the promoted sorbent to have both high capacity and high reactivity toward mercury. The promoted sorbent can be utilized in a very finely powdered form to minimize mass-transfer limitations. However, again, the reactivity should be very high to capture all of the mercury encountered by the fine particles. Additionally, use of these enhancement technologies allows capture to be effective for larger sorbent particles which also allows separation of the promoted sorbent from the ash to enable subsequent regeneration as well as ash utilization. One feature of this invention is the process to prepare a promoted sorbent containing a halide component formed on and/or within the base sorbent structure that provides a sorbent that is highly active on initial contact with the mercury-contaminated gas stream, which allows for very effective capture of the mercury.

[0087] The inventive sorbents chemically combine bromine species with Lewis acid/basic sites on the base sorbent. For example, x-ray photoelectron spectroscopy has established that the addition of bromine, chlorine, HBr, or HCl formed a chemical compound in the carbon structure. Thus the promoted sorbent produced from halogen and base sorbent does not represent a molecular halogen form, but rather a new chemically modified structure. This phenomenon may not occur with the less reactive iodine, where an I_2 molecular complex can exist on the carbon basal plane. In the case of bromine, modified cationic carbon has a high chemical potential for oxidation of

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mercury. Thus an entirely new model is presented for the reactivity of the bromine-treated carbon with mercury shown in Figure 2. The reactive carbon form can preferably be generated by the addition of bromine, hydrogen bromide, or combinations of bromine and other elements, as described herein. Halogen treatment resulted in higher-activity carbons because the halide anions (especially bromide and iodide) were effective in promoting oxidation by stabilizing the developing positive charge on the mercury in the transition state for oxidation. Based on this model, several innovative, inexpensive, activity-enhancing features have been developed.

Optional Second Component

[0088] It has been demonstrated that addition of an optional second component, in addition to the bromine, results in improved reactivity and capacity for the promoted sorbent, typically exceeding that of both the untreated base sorbent and the brominated carbon. The second compound comprises either a second halogen or a compound derived from a second halogen, such as HI. Thus in addition to having a reactive carbon form present, the second component generates a Lewis base with greater ability to stabilize the developing positive charge on the mercury. Thus the second component is an element with more polarized electrons (4p and 5p).

Optional Alkali Component

[0089] It has been demonstrated that addition of an optional alkali component with a base or promoted activated carbon results in improved mercury capture, typically exceeding that of both the untreated carbon and the promoted carbon. Test data indicate that flue gas contaminants, flue gas constituents (SO_2, NO_x, HCl, etc), operating temperature, mercury form, and mercury concentration may impact the effectiveness of the alkali addition. This suggests the need to be able to adjust and tailor the alkali-to-activated-carbon ratio on-site in order to overcome and optimize for a given set of site conditions.

[0090] The synergy that can be gained when coinjecting the two materials can be explained as follows. First, testing shows that binding sites on activated carbon (hereinafter "AC") can be consumed by chlorine species, sulfur species (i.e., sulfates), and other flue gas contaminants (arsenates, selenates, etc). The addition of optional alkali material will interact and react with these species/contaminants, thus minimizing their consumption of AC mercury binding sites. Second, testing also shows that standard AC will continue to oxidize mercury, even though the binding sites are fully

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consumed. This oxidized mercury can then react with alkali material and subsequently be captured by particulate control devices. Consequently, the addition of the optional alkali component acts to protect mercury-binding sites and capture oxidized mercury, thereby resulting in improved mercury reduction at lower cost. Alkali is generally much lower in cost (~ an order of magnitude less) than AC; thus more of it can be used, still resulting in overall lower costs.

In-Flight Sorbent Preparation

[0091] As stated previously, the halogen promoted sorbent can be readily produced in-flight. This is accomplished by, for example, contacting the vapors of any combination of halogens and, optionally, a second component, in-flight, with base sorbent particles. The particles may be dispersed in a stream of transport air (or other gas, such as the flue gas itself), which also conveys the halogen/halide-promoted sorbent particles to the flue gas duct, or other contaminated gas stream, from which mercury is to then be removed. There is no particular temperature requirement for this contact. This technology is obviously very simple to implement, and results in a great cost savings to facilities using this technology for mercury capture.

[0092] Referring to Figures 5A and 5B, process flow diagrams are shown as examples of the process for mercury removal from a mercury containing gas, such as a flue gas. In an embodiment shown at Figure 5A, gas inlet stream 501 passes through chamber 301 and enters air heater 302 as stream 502; then it exits air heater 302 as stream 503 and passes through particulate control device 303 and enters scrubber 304 as stream 504; finally it exits scrubber 304 as stream 505. As shown, chamber 301 is a boiler, however, one skilled in the art will appreciate that chamber 301 can also be the combustion chamber of a coal fired boiler, a stand alone combustion chamber or any other chamber in which mercury containing gas is either generated or passed through. Injection streams 401, 402, 403 and 404, in addition to stream 501, are multiple locations wherein promoter, base sorbent, or a combination of promoter and base sorbent may be introduced.

[0093] Figure 5B shows an embodiment similar to that of FIG. A, but with the position of the scrubber and particulate control device switched. Specifically, gas inlet stream 501' passes through chamber 301' and enters air heater 302' as stream 502'; then it exits air heater 302' as stream 503' and passes through scrubber 304' and enters particulate control device 303' as stream 504'; finally it exits particulate control device 303' as stream 505'. As shown, chamber 301 is a boiler, however, one skilled in the art will

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appreciate that chamber 301 can also be the combustion chamber of a coal fired boiler, a stand alone combustion chamber or any other chamber in which mercury containing gas is either generated or passed through. Injection stream locations 401', 402', 403' and 404', in addition to stream 501', represent multiple locations wherein promoter, base sorbent, or a combination of promoter and base sorbent may be introduced.

[0094] At each of the aforementioned injection stream locations 401, 401', 402, 402', 403, 403', 404 and 404', 501, 501' multiple injection points are contemplated so that promoter and base sorbent may be injected as a single injection stream or as separate injection streams, as further illustrated in the following exemplary embodiments.

[0095] In an embodiment, a promoter is introduced to chamber 301 at location 401. A base sorbent (carbon, non-carbon, or their combination) is introduced at location 402 upstream of air heater 302. In a further embodiment, a promoter is introduced to at location 402 upstream of air heater 302. A base sorbent (carbon, non-carbon, or their combination) is also introduced at location 402 either as a separate stream or as a mixed single stream with the promoter. In another embodiment, a promoter is introduced at locations 401 and 402. A base sorbent (carbon, non-carbon, or their combination) is introduced at location 402 either as a separate stream or as a mixed single stream with the promoter.

[0096] With the aid of this disclosure, one of ordinary skill in the art will be able to configure the process in many different fashions for mercury removal using the promoted sorbent. All these configurations are considered equivalents of the disclosed process and therefore are within the scope of the claimed invention.

Advantages of On-Site Preparation

[0097] In-flight preparation of the halogen/halide-promoted sorbent on location produces certain advantages. For example, the treatment system can be combined with the base sorbent injection system at the end-use site. With this technique, the halogen/halide is introduced to the base sorbent air (or other gas, including to the flue gas or other mercury containing gas) mixture in a transport line (or flue gas duct part of the base sorbent storage and injection system). This provides the following benefits over current conventional concepts for treating sorbents off-site:

- Capital equipment costs at a treatment facility are eliminated.
- Costs to operate the treatment facility are eliminated.
- There are no costs for transporting carbon and additive to a treatment facility.

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- The inventive process uses existing hardware and operation procedures.
- The inventive technology ensures that the sorbent is always fresh and, thus, more reactive.
- No new handling concerns are introduced.
- There are no costs for removing carbon from treatment system.
- The inventive process allows rapid on-site tailoring of additive–sorbent ratios in order to match the requirements of flue gas changes, such as may be needed when fuels are changed or loads are reduced, thus further optimizing the economics.
- The inventive technology reduces the amount of spent sorbents that are disposed.

[0098] With the foregoing and other features in view, there is provided, in accordance with the present invention, embodiments including a process for preparing and regenerating halogen/halide-promoted sorbents, whose activity for mercury capture is enhanced by the addition of halogen (e.g., bromine) to the base sorbent structure.

Sorbent Injection Location

[0099] Some of the preferred embodiments contemplate the use of a halogen-promoted sorbent in a powdered form that has been injected into a flue gas stream before or after ash particulates have been removed. Other embodiments of the inventive composition of the halogen-promoted sorbent comprise a powdered modified AC prepared by adding Br_2 or HBr plus a second optional component. Other embodiments allow the addition of the optional alkali component in conjunction with a base AC and/or with the use of a halogen-based sorbent and any other combinations of the sorbent technologies provided in this patent. Other embodiments allow for in-flight preparation of promoted sorbents by using and combining the promoters and base sorbents. Alternatively, embodiments include methods wherein the base sorbent is on a moving contactor consisting of particles or fibers containing one or more of the compositions listed above.

Sorbent Regeneration

[00100] Any of the above embodiments of the halogen/halide-promoted carbon and/or non-carbon sorbent can be easily regenerated; the poisoning contaminants from the flue gas are preferably removed, and an inexpensive promoting agent added, to restore mercury sorption activity. This process of promoting the activity of the carbon and/or non-carbon itself contrasts with the earlier, more expensive, conventional methods of

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adding a reagent (such as peroxide, gold, triiodide, etc.) to a base sorbent. The halogen/halide-promoted carbon sorbent of the present invention, treated with bromine and/or optional components, is noncorrosive. Detailed examples of sorbent regeneration techniques are described in copending, commonly owned PCT Patent Application No. PCT/US04/12828, titled "PROCESS FOR REGENERATING A SPENT SORBENT".

Sorbent Injection Control Schemes

[00101] Another advantage of the present invention relates to the use of a feedback system to more efficiently utilize certain aspects of the invention. Where possible and desirable, the mercury control technology of the present invention may preferably utilize continuous measurement of mercury emissions as feedback to assist in control of the sorbent injection rate. Tighter control on the sorbent and optional component(s) levels can be achieved in this way, which will ensure mercury removal requirements are met with minimal material requirements, thus minimizing the associated costs. In an embodiment, the mercury emissions are continuously measured downstream of the injection location, preferably in the exhaust gas at the stack.

EXAMPLES

[00102] To more clearly illustrate the present invention, an example is presented below. This example is intended to be illustrative, and no limitations to the present invention should be drawn or inferred from the example presented herein.

Example 1 - Non-carbon-Promoted Sorbent Tests at Full-Scale Plants

[00103] Full-scale commercial tests were conducted at several coal-burning facilities equipped with ESPs wherein three types of pre-cursers were injected upstream of an ESP at 325^{0}F. In all, more than thirty tests were performed with variations in base sorbent material, base sorbent material injection rates and promoter injection rates. Promoters were halogen-based materials. Base sorbents were clay-based materials such as perlite and pumacite, which are vesicular forms of tectosilicates comprising silicates and aluminate tetrahedra with alkali metals in the interstitual spaces. Each base sorbent was injected upstream from the ESP.

[00104] The flue gas flow rate for the test was approximately 23 million dsft^{3}/hr with a corresponding mercury flow rate of 0.0097 lbs Hg/hr. Initial mercury concentration in

the flue gas prior to the introduction of the base sorbent or promoted sorbent was approximately 0.000417 lbs Hg/million dsft^3.

[00105] As shown in Table 1, each base sorbent was injected at three different test rates: about 100 lb/hr ("Low"), about 150 lb/hr ("Ave.") and about 200 lb/hr ("High"). At each test rate, the mercury removal rate was measured both with and without the use of a promoter to show the relative benefit of introducing the promoter into the flue gas stream. For the tests where promoter was injected into the flue gas stream, promoter was injected at about 20 lb/hr for the "Low" tests, at about 25 lb/hr for the "Ave." tests and from about 30 to about 50 lb/hr for the "High" tests. The percent mercury removal for each test was calculated based on measurements taken from the inlet flue gas and the outlet flue gas. The results as shown in Table 1 clearly show a significant improvement in mercury capture when a promoter and base sorbent are injected, as compared to the injection of a base sorbent alone.

Table 1. Test Data for Non Carbon-Promoted Sorbents

Base Sorbent Material	Base Sorbent Injection Rate (lb/hr)			Promoter Injection Rate (lb/hr)			Percent Removal of Mercury (%)		
	Low	Ave.	High	Low	Ave.	High	Low	Ave.	High
Perlite	100	150	200	0	0	0	37	40	43
	100	150	200	20	25	35	55	72	74
Punacite	100	150	200	0	0	0	25	25	25
	100	150	200	20	25	50	52	60	65
Clay-Based Mix of Silica and Alumina	100	150	200	0	0	0	50	52	54
	100	150	200	20	25	30	58	65	74

[00106] While the preferred embodiments of the invention have been shown and described, modifications thereof can be made by one skilled in the art without departing from the spirit and teachings of the invention. The embodiments described herein are exemplary only and are not intended to be limiting. Many variations and modifications of the invention disclosed herein are possible and are within the scope of the invention.

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Accordingly, the scope of protection is not limited by the description set out above, but is only limited by the claims which follow, that scope including all equivalents of the subject matter of the claims.

[00107] The examples provided in the disclosure are presented for illustration and explanation purposes only and are not intended to limit the claims or embodiment of this invention. While the preferred embodiments of the invention have been shown and described, modifications thereof can be made by one skilled in the art. Process criteria, equipment, and the like for any given implementation of the invention will be readily ascertainable to one of skill in the art based upon the disclosure herein. The embodiments described herein are exemplary only and are not intended to be limiting. Many variations and modifications of the invention disclosed herein are possible and are within the scope of the invention. Use of the term "optionally" with respect to any element of the invention is intended to mean that the subject element is required, or alternatively, is not required. Both alternatives are intended to be within the scope of the invention.

[00108] The discussion of a reference in the Background is not an admission that it is prior art to the present invention, especially any reference that may have a publication date after the priority date of this application.

[00109] Although the invention is described herein as a promoted sorbent material and associated processes for its preparation and use, it is nevertheless not intended to be limited to the details described.

WHAT IS CLAIMED IS:

1. A method for separating mercury from a mercury containing gas comprising:

(a) providing a carbon sorbent material and a non-carbon sorbent material, wherein the carbon sorbent material comprises activated carbon;

(b) providing a halogen or halide promoter;

(c) promoting at least a portion of the carbon sorbent material and the non-carbon sorbent material by chemically reacting the carbon sorbent material and the non-carbon sorbent material with the halogen or halide promoter to form a promoted halogenated carbon sorbent and a promoted halogenated non-carbon sorbent;

(d) chemically reacting elemental mercury in the mercury containing gas with the promoted halogenated carbon sorbent and the promoted halogenated non-carbon sorbent to form a mercury/sorbent chemical composition; and

(e) separating particulates from the mercury containing gas to form a cleaned gas, the particulates including ash and the mercury/sorbent chemical composition.

2. The method of claim 1, further comprising the step of injecting the non-carbon sorbent material and the halogen or halide promoter into the mercury containing gas.

3. The method of claim 2, wherein said halogen or halide promoter and the non-carbon sorbent material are injected into the mercury containing gas at the same location.

4. The method of claim 2, wherein said halogen or halide promoter and the non-carbon sorbent material are injected into the mercury containing gas at separate locations.

5. The method of claim 4, wherein said halogen or halide promoter is injected into the mercury-containing gas upstream of the injection of said non-carbon sorbent material.

6. The method of claim 5, wherein said halogen or halide promoter is injected into a combustion chamber that produces a mercury-containing gas, and the non-carbon sorbent material is injected downstream of the combustion chamber.

7. The method of claim 6, wherein said halogen or halide promoter is additionally injected downstream of the combustion chamber.

8. The method of claim 6, wherein said combustion chamber is a boiler and the mercury-containing gas is a flue gas.

9. The method of claim 3, wherein said halogen or halide promoter and non-carbon sorbent material are injected downstream of a chamber that produces a mercury-containing gas.

10. The method of claim 9, wherein said chamber is a boiler and the mercury-containing gas is a flue gas.

11. The method of claim 2, wherein the rate at which said non-carbon sorbent material is injected or the rate at which said promoter is injected or combination thereof is adjusted according to a monitored mercury content in the cleaned gas so that the mercury content of the cleaned gas is maintained at a desired level.

12. The method of claim 1, wherein said non-carbon sorbent material is selected from the group consisting of porous felsic materials, vesicular felsic materials, porous basaltic materials, vesicular basaltic materials, clay-based compounds, alkaline compounds, calcium hydroxide compounds, sodium acetate compounds, bicarbonate compounds, and combinations thereof.

13. The method of claim 12, wherein said non-carbon sorbent material is a material that reacts with oxidized mercury in the mercury-containing gas to form a second mercury/sorbent chemical composition.

14. The method of claim 1, wherein said non-carbon sorbent material comprises Lewis basic groups.

15. The method of claim 12, wherein said non-carbon sorbent material comprises amorphous forms of tectosilicates comprising nanoscale cavities lined with Lewis basic oxygen associated with alkaline-earth metals.

16. The method of claim 15, wherein said alkaline-earth metals comprise Group I and Group II alkaline-earth metals.

17. The method of claim 1 wherein said non-carbon sorbent material comprises amorphous forms of phyllosilicates comprising nanoscale cavities lined with Lewis basic oxygen.

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18. The method of claim 1, wherein said promoted non-carbon sorbent comprises metastable complexes formed between said promoter and inorganic species on the non-carbon sorbent.

19. The method of claim 18, wherein said inorganic species is selected from the group consisting of sodium compounds, calcium compounds, magnesium compounds, aluminum compounds, iron compounds, and combinations thereof.

20. The method of claim 1, wherein said promoted non-carbon sorbent comprises metastable complexes formed between said promoter and metal-oxygen-metal structures on the non-carbon sorbent.

21. The method of claim 20, wherein said promoter after being complexed with the metal-oxygen-metal structures is in the form selected from the group consisting of a dihalogen group, a halogen atom, a hydrohalogen group, a Group V halide, a Group VI halide, and combinations thereof.

22. The method of claim 1, wherein said promoted non-carbon sorbent comprises activated Lewis basic groups or activated Lewis acid groups or combination thereof.

23. The method of claim 1, wherein said interaction between promoted non-carbon sorbent and said mercury-containing gas stream comprises:

mercury diffusing from the gas phase onto a surface of said non-carbon promoted sorbent; and

reacting with activated Lewis basic groups or activated Lewis acid groups or combination thereof to cause chemisorption on a sorbent surface.

24. The method of claim 1 further comprising pretreating said non-carbon sorbent to increase the number of Lewis basic groups or Lewis acid groups or combination thereof on said sorbent.

25. The method of claim 24, wherein pretreating said non-carbon sorbent comprises chemical treatment, thermal treatment, vacuum treatment, and combinations thereof.

26. The method of claim 25, wherein said chemical treatment comprises acid treatment and alkaline treatment

27. The method of claim 1 further comprising introducing an alkali component into the mercury-containing gas.

28. The method of claim 1, wherein said carbon sorbent or said promoter or combination thereof are introduced into the mercury-containing gas at one or more locations.

29. The method of claim 28, wherein the rate at which said carbon sorbent is introduced or the rate at which said promoter is introduced or combination thereof is adjusted according to a monitored mercury content in the cleaned gas so that the mercury content of the cleaned gas is maintained at a desired level.

30. The method of claim 1, where the promoted halogenated carbon sorbent and the promoted halogenated non-carbon sorbent comprise 1 to 30 grams of the halogen or halide promoter per 100 grams of the carbon sorbent and the non-carbon sorbent.

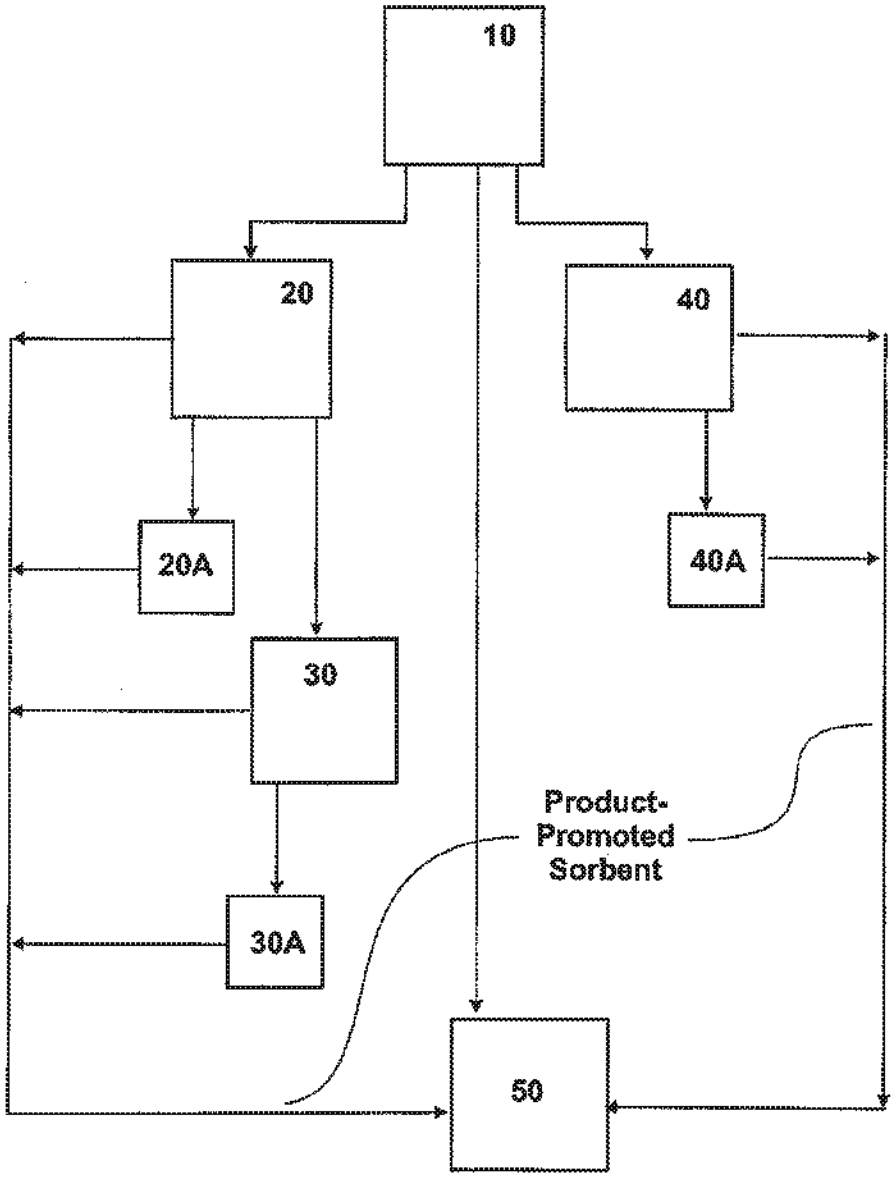


FIG. 1

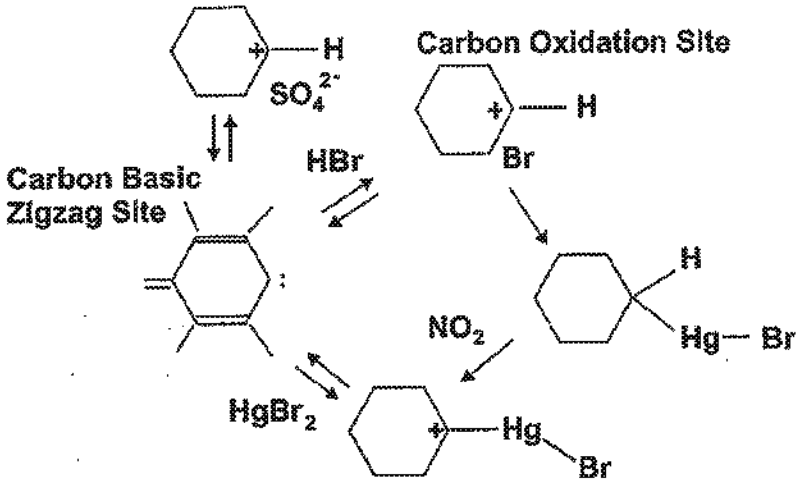

Carbon Oxidation Site
H
SO_4^{2-}
H
Br
HBr
Carbon Basic
Zigzag Site
H
Hg— Br
NO_2
$HgBr_2$
Hg
Br

FIG. 2

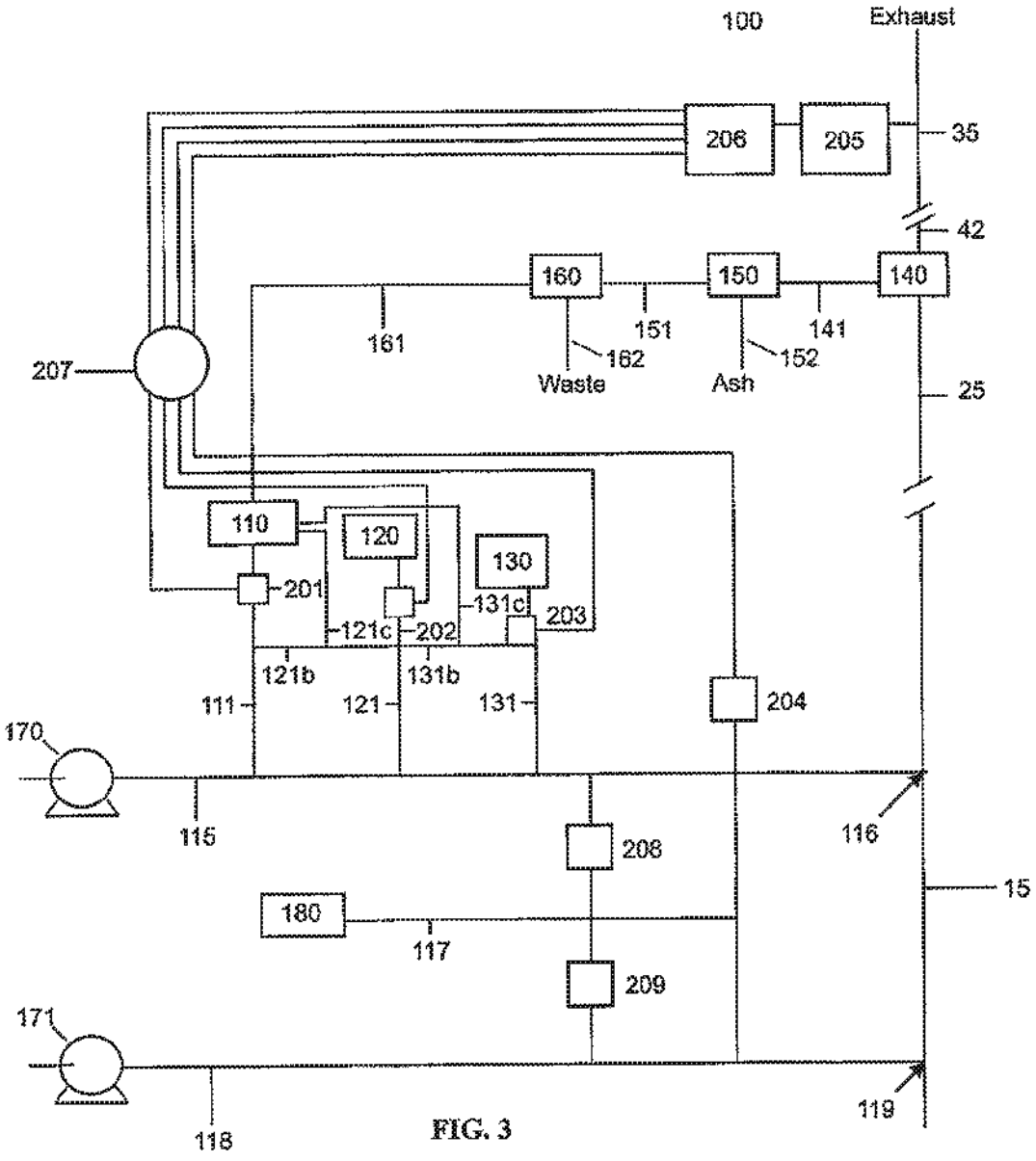

100
Exhaust
206
205
35
42
160
150
140
161
151
141
162
152
Waste
Ash
207
25
110
120
130
201
131c
203
121c
202
121b
131b
111
121
131
204
170
115
116
208
15
180
117
209
171
118
119

FIG. 3

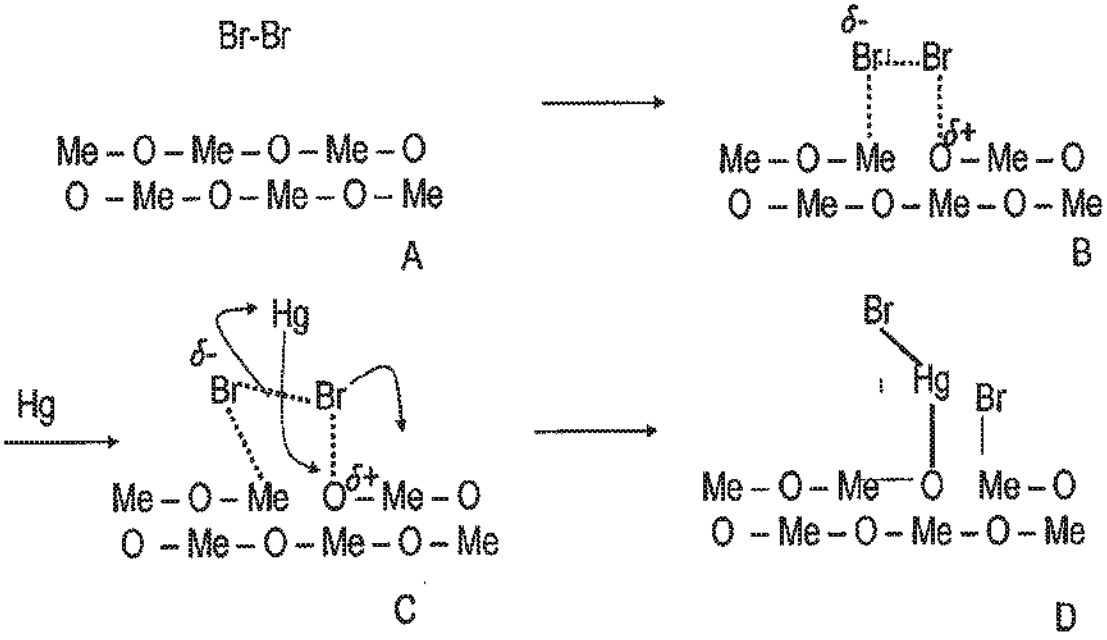

Br-Br
Me – O – Me – O – Me – O
O – Me – O – Me – O – Me
A
δ-
Br
Br
δ+
Me – O – Me
O – Me – O
Me – O – Me – O – Me
B
Hg
Hg
δ-
Br
Br
δ+
Me – O – Me
O – Me – O
Me – O – Me – O – Me
C
Br
Hg
Br
Me – O – Me
O
Me – O
O – Me – O – Me – O – Me
D

FIG. 4

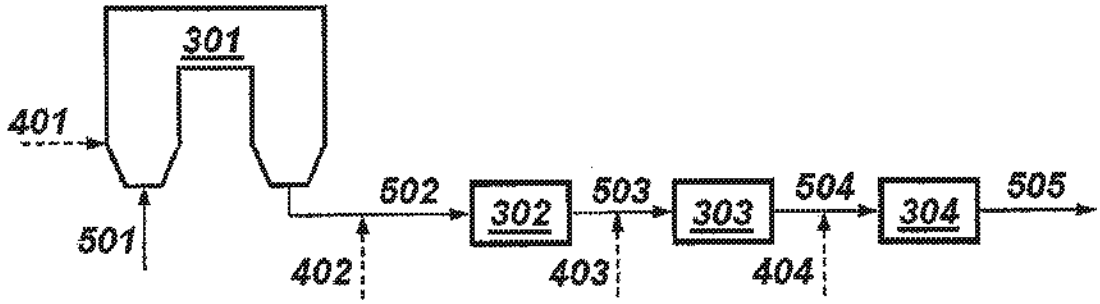


FIG. 5A

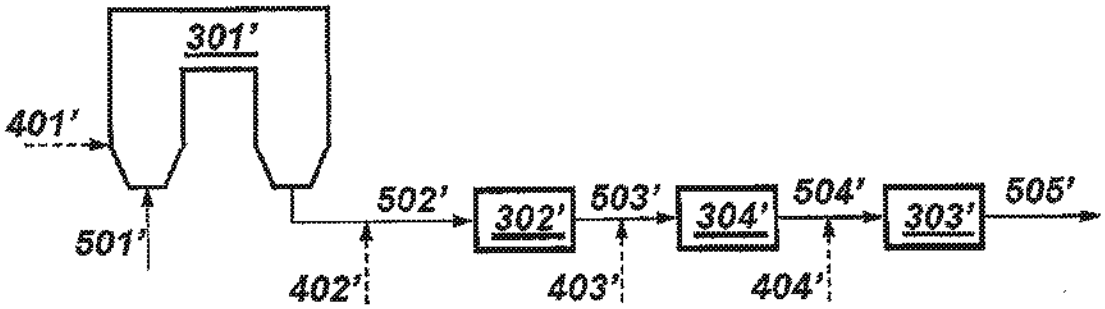


FIG. 5B

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Espacenet

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Sorbents for the oxidation and removal of mercury

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Applicant(s): ENERGY & ENVIRON RES CT FOUND [US] ± (ENERGY & ENVIRONMENTAL RES CT)

Classification: - international: ***B01D53/02***
- cooperative: **B01D53/10; B01D53/64; B01J20/20; B01J20/22;** B01D2253/102; B01D2257/602; Y10S95/901

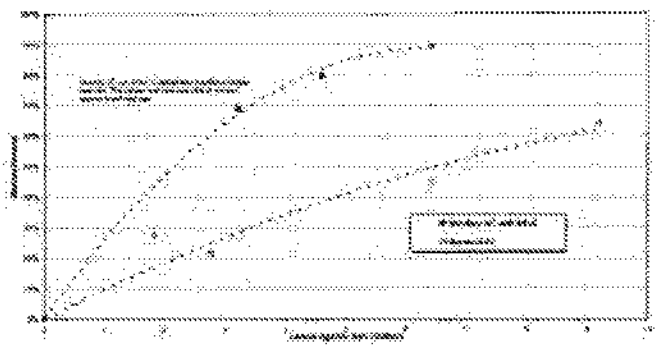
Application number: CN2005837037 20050823

Priority number(s): WO2005US30018 20050823 ; US20040605640P 20040830 ; US20050209163 20050822

Also published as: CN101048218 (B) CA2584327 (A1) CA2584327 (C) EP1796816 (A2) EP1796816 (A4) more

Abstract of CN101048218 (A)

A promoted activated carbon sorbent is described that is highly effective for the removal of mercury from flue gas streams. The sorbent comprises a new modified carbon form containing reactive forms of halogen and halides. Optional components may be added to increase reactivity and mercury capacity. These may be added directly with the sorbent, or to the flue gas to enhance sorbent performance and/or mercury capture. Mercury removal efficiencies obtained exceed conventional methods. The sorbent can be regenerated and reused. Sorbent treatment and preparation methods are also described. New methods for in-flight preparation, introduction, and control of the active sorbent into the mercury contaminated



gas stream are described.

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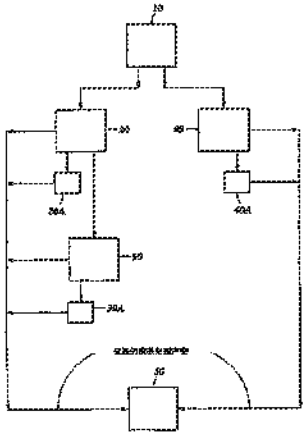
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权利要求书 6 页 说明书 32 页 附图 7 页

[54] 发明名称

用于汞的氧化及清除的吸附剂

[57] 摘要

本发明描述了一种促进的活性炭吸附剂，其用于从烟道气流中高效地清除汞。该吸附剂包括含有活性形式的卤素和卤化物的新改性的碳形式。可以加入任选的组分以提高活性和汞容量。可以将这些任选组分与吸附剂直接加入，或者将这些加入到烟道气中以提高吸附剂性能和/或汞的捕集。得到的汞清除效率超过了常规的方法。吸附剂可以再生及重新使用。还描述了吸附剂处理及制备方法。描述了动态制备、引入及控制活性吸附剂到汞污染的气流中的新方法。



1. 一种促进的碳吸附剂，其包含已与选自卤化物、卤素及其组合的促进剂反应的基体活性碳，从而使反应产物有效用于从气流中清除汞。

2. 权利要求 1 的促进的碳吸附剂，其中基体活性碳选自粉状活性碳、粒状活性碳、碳黑、碳纤维、气凝胶碳、热解炭、平均粒度大于 40 微米的活性碳及其组合，促进剂选自分子态卤素、V 族卤化物、VI 族卤化物、氢卤化物及其组合。

3. 权利要求 2 的促进的碳吸附剂，其中相对于每 100 克的基体活性碳，该吸附剂包含约 1 至约 30 克的促进剂。

4. 权利要求 3 的促进的碳吸附剂，还包括含有卤素或氢卤化物的任选的次级组分，从而提高吸附剂的活性和汞容量。

5. 权利要求 4 的促进的碳吸附剂，还包含选自碱元素、碱土元素、碱盐、碱土盐及其组合的任选的碱性组分。

6. 权利要求 4 的促进的碳吸附剂，其中成品吸附剂中任选的次级组分的浓度范围为成品吸附剂中促进剂浓度的约 1 至约 15 wt%。

7. 权利要求 6 的促进的碳吸附剂，其中任选的次级组分选自 V 族卤化物、VI 族卤化物、HI、HBr、HCl 及其组合。

8. 权利要求 7 的促进的碳吸附剂，其中任选的次级组分选自 I_2、HI 及其组合。

9. 权利要求 8 的促进的碳吸附剂，其中与基体活性碳结合时的促

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进剂基本上是蒸汽形式的。

10. 权利要求 8 的促进的碳吸附剂，其中促进剂在与基体活性碳反应之前与有机溶剂结合。

11. 权利要求 8 的促进的碳吸附剂，其中促进剂和任选的次级组分基本上同时与基体活性碳结合。

12. 权利要求 8 的促进的碳吸附剂，还包含选自 S、Se、H_2S、SO_2、H_2Se、SeO_2、CS_2、P_2S_5 及其组合的汞稳定剂。

13. 权利要求 12 的促进的碳吸附剂，其中促进剂选自 Br_2、HBr 及其组合。

14. 权利要求 3 的促进的碳吸附剂，还包含选自 S、Se、H_2S、SO_2、H_2Se、SeO_2、CS_2、P_2S_5 及其组合的汞稳定剂。

15. 权利要求 14 的促进的碳吸附剂，还包括含碱或碱土元素的任选的碱性组分，其中促进剂选自 Br_2、HBr 及其组合。

16. 一种方法，其包括：提供粒状的活性碳；使活性碳与选自卤素、卤化物及其组合的促进剂反应，从而使反应产物包含有效用于从气流中清除汞的促进的碳吸附剂。

17. 权利要求 16 的方法，其中相对于每 100 克的活性碳，反应产物包含约 1 至约 30 克的促进剂。

18. 权利要求 16 的方法，其中反应产物的平均粒度分布大于约 40 微米。

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19. 权利要求 16 的方法，其中促进剂选自分子态卤素、氢卤化物、V 族卤化物、VI 族卤化物及其组合。

20. 权利要求 19 的方法，其中促进剂与活性碳接触时是在气相中。

21. 权利要求 19 的方法，其中促进剂在接触活性碳时在有机溶剂中。

22. 权利要求 20 的方法，其中促进剂选自 Br_2、V 族溴化物、VI 族溴化物及其组合。

23. 权利要求 18 的方法，还包括提供任选的碱性组分以改善汞清除的有效性。

24. 权利要求 20 的方法，还包括使粒状的活性碳与包括卤素或氢卤化物的任选的次级组分进行反应，从而使吸附剂的活性和汞容量得以提高。

25. 权利要求 24 的方法，还包括提供任选的碱性组分以改善汞清除的有效性。

26. 权利要求 24 的方法，其中促进剂和任选的次级组分同时与活性碳接触。

27. 权利要求 24 的方法，其中任选的次级组分包括 I_2 或 HI。

28. 权利要求 20 的方法，还包括加入选自 S、Se、H_2S、SO_2、H_2Se、SeO_2、CS_2、P_2S_5 及其组合的汞稳定剂。

29. 权利要求 24 的方法，还包括加入选自 S、Se、H_2S、SO_2、H_2Se、

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SeO_2、CS_2、P_2S_5及其组合的汞稳定剂。

30. 一种降低烟道气中的汞的方法，其包括：提供吸附剂，将吸附剂注入到包含汞的烟道气流中，在吸附剂上收集烟道气中超过70wt%的汞以产生清洁的烟道气，以及从清洁的烟道气中基本回收吸附剂。

31. 权利要求30的方法，还包括将任选的碱性组分共注入到含汞的烟道气流中。

32. 权利要求30的方法，还包括监测清洁的烟道气中的汞含量、使回收的吸附剂再生以及应用清洁的烟道气中的监测汞含量来控制吸附剂的注入速率。

33. 权利要求30的方法，其中通过使活性碳与促进剂在气动运输管线内反应来动态地制备注入的吸附剂，反应产物从该管线中注入到含汞的烟道气流中。

34. 权利要求33的方法，其中促进剂选自分子态卤素、卤化物及其组合。

35. 权利要求34的方法，其中促进剂选自Br_2、HBr、V族溴化物、VI族溴化物及其组合。

36. 权利要求35的方法，其中促进剂在气相中或以蒸汽形式进行反应。

37. 权利要求36的方法，其中相对于每100克的活性碳加入约1至约30克促进剂。

38. 权利要求37的方法，其中通过在气动运输管线内使活性碳、

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促进剂和任选的次级组分进行反应来动态地制备注入的吸附剂，从而提高吸附剂的活性和容量，反应产物从该管线中注入到包含汞的烟道气流中。

39. 权利要求 38 的方法，其中任选的次级组分选自碘、氢卤化物、V 族卤化物、VI 族卤化物及其组合。

40. 权利要求 39 的方法，还包括将任选的碱性组分共注入到含汞的烟道气流中。

41. 权利要求 39 的方法，其中任选的次级组分的加入量为促进剂含量的约 1 至约 15 wt%。

42. 权利要求 41 的方法，其中任选的次级组分选自 I_2、HI 及其组合。

43. 权利要求 42 的方法，其中在气相中或以蒸汽形式加入任选的次级组分。

44. 权利要求 41 的方法，还包括在吸附剂中加入选自 S、Se、H_2S、SO_2、H_2Se、SeO_2、CS_2、P_2S_5 及其组合的汞稳定剂。

45. 权利要求 32 的方法，还包括应用清洁的烟道气中的监测汞含量来控制吸附剂的组成。

46. 权利要求 40 的方法，还包括监测清洁的烟道气中的汞含量以及应用监测的汞含量来控制吸附剂的组成和碱与吸附剂的注入比。

47. 权利要求 45 的方法，其中通过使活性碳与促进剂在气动运输管线内反应来动态地制备注入的吸附剂，反应产物从该管线注入到含

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汞的烟道气流中，其中促进剂选自分子态卤素、卤化物及其组合，其中促进剂在气相中或以蒸汽形式进行反应，其中相对于每 100 克的活性碳加入约 1 至约 30 克促进剂，其中至少部分地基于清洁的烟道气中的监测汞含量、通过数字计算机来确定促进剂的加入速率和吸附剂的注入速率。

48. 一种降低气流中的汞含量的方法，其包括以下步骤：将质均粒度>40μm 的粉状活性碳吸附剂注入到包含汞及灰分粒子的气流中，从气流中将汞捕集于吸附剂粒子上，将吸附剂粒子与灰分粒子机械分离开，以及将分离的吸附剂粒子再注入到气流中。

49. 权利要求 48 的方法，还包括使吸附剂粒子再生，从而从中基本清除污染物以恢复汞捕集的容量。

50. 权利要求 48 的方法，还包括将选自碱元素、碱土元素、碱盐、碱土盐及其组合的碱性组分共注入到气流中。

51. 权利要求 48 的方法，其中粉状活性碳吸附剂还包含选自卤化物、卤素及其组合的促进剂。

52. 权利要求 51 的方法，还包括将选自碱元素、碱土元素、碱盐、碱土盐及其组合的碱性组分共注入到气流中。

53. 权利要求 48 的方法，其中微粒活性碳吸附剂具有的质均粒度>60μm。

54. 权利要求 52 的方法，其中微粒活性碳吸附剂具有的质均粒度>60μm。

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用于汞的氧化及清除的吸附剂

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发明背景

发明技术领域

本发明涉及用于从气化系统的产物气或者烟道气中清除污染物的方法及材料。具体而言，本发明涉及通过高活性的可再生吸附剂从化石燃料的燃烧或气化期间产生的气流中清除汞。

发明背景

诸如煤炭的化石燃料的燃烧和气化能产生包含源自于燃料的汞及其它痕量元素的烟道气。必须通过使用吸附剂、涤气器、过滤器、沉降器以及其它清除技术来控制汞(和其它污染物)向环境中的排放。在燃烧和气化期间汞最初以元素态的形式存在。在诸如燃烧系统的管道及烟道的下游工艺部分中，一些元素态的汞被氧化。被氧化的量取决于烟道气中酸性气体的存在量以及其它的因素。汞的量随燃料不同而变化，但煤炭燃烧的烟道气流中汞的浓度通常为每十亿份中低于 5 份(ppb)。诸如电力设施的大型煤炭燃烧设备每天可能排出一磅的汞或更多。汞清除的应用领域包括但不限于煤炭(或其它化石燃料)燃烧中的烟道气、废物焚烧、气化中的产物气以及矿物加工、金属提炼、干馏、水泥制造、氯碱工厂、牙科设施和火化场中的尾气。

汞吸附剂技术

已经研究了几种类型的用于烟道气中汞的控制方法，包括将细吸附剂粒子注入到烟道气管道内，并使烟道气通过吸附剂床。细粒子注

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入吸附剂包括活性碳、金属氧化物吸附剂、硫化钠粒子和碱性硅酸盐或氧化物吸附剂。当采用粒子注入时，在袋滤室或静电沉降器(ESP)中将捕集到吸附剂粒子上的汞从气流中清除并与灰分微粒一起收集起来。硫化物和碱性硅酸盐及氧化物粒子仅对氧化态的汞是有效的，且与碳粒子相比，金属氧化物吸附剂显示出降低的捕集动力学性能。此外，将细碳粒子注入到烟道气流内来清除汞，尤其是清除元素态的汞，仅仅是部分成功，在使用 FF(织物过滤器)以收集碳和灰分的某些应用中，仅仅达到约 60%的有效清除。当使用 ESP 来收集碳时，因为碳与气体的接触时间非常短，观察到得到甚至更低的清除比率。

现有的碳注入系统的主要问题是，吸附剂初始为非活性的，只有在烟道气中长时间地暴露以后吸附剂才能具备有效的适应性，并提供对气体中汞的增强活性。因此，为了有效地捕集汞，必须以较大的量、较高的吸附剂/汞比例使用这些吸附剂。这些吸附剂往往相对昂贵，不易从灰分中分离出来以再生及重新使用。灰分中碳的收集也造成了固体废物处理的问题，用过的吸附剂可能污染收集的灰分，妨碍其在各种应用当中的用途。

因此，需要有一种更加经济和有效的汞清除技术。本发明提供包含汞的污染物的成本有效清除方法，其使用吸附剂增强添加剂和/或高活性吸附剂，接触时间为若干秒(或更短)，且可以再生及重新使用。

发明概述

因此，本发明的目的是克服现有技术的不足，从而提供新的和经济的方法，用于在利用化石燃料而产生的气体中清除汞。

本发明描述了一种卤素/卤化物促进的活性碳吸附剂，其用于高效地从烟道气流中清除汞。该吸附剂含有新的卤化物改性碳形式，其包含由溴(或卤化物或其它卤素)与碳反应生成的活性化合物。可以加入任选的次级组分和碱以进一步提高活性及汞容量。得到的汞清除效率超

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过或相当于常规的方法，还具有附加的益处例如成本降低。任选地，可以再生及重新使用吸附剂。还描述了吸附剂处理和/或制备方法。描述了活性吸附剂的动态(in-flight)制备、引入到汞污染的气流中以及控制的新方法。

在一些实施方案中提供了促进的碳吸附剂，其包含已经与选自卤化物、卤素及其组合的促进剂反应的基体活性碳，从而使反应产物可有效用于从气流中清除汞。

在一个实施方案中提供了促进的碳吸附剂，其中基体活性碳选自粉状活性碳、粒状活性碳、碳黑、碳纤维、气凝胶碳、热解炭、平均粒度大于所产生的飞灰的活性碳(从而可以从中物理地分离出来) 以及它们的组合。促进剂选自分子态卤素、第 V 族(全部采用 CAS 术语)卤化物、第 VI 族卤化物、氢卤化物以及它们的组合。在一个实施方案中，基体活性碳可以具有这样的质均粒径，使得可以基本上通过物理方法将其从气流中夹带的含有欲清除的汞的灰分中分离出来。在一个实施方案中，基体活性碳可以具有大于约 40 微米的质均粒径。

在另一个实施方案中，对于每 100 克的基体活性碳而言，吸附剂包含约 1 至约 30 克的促进剂。另一实施方案中还包含任选的包括卤素或氢卤化物的次级组分，从而提高吸附剂的活性和汞容量。

在另一个实施方案中，成品吸附剂中任选次级组分的浓度范围在成品吸附剂中促进剂浓度的约 1 至约 15 wt%以内。

在另一个实施方案中，优选可以加入任选的碱性组分以通过该碱与初级吸附剂的组合提供协同效应。

在另一个实施方案中，任选的次级组分选自 V 族卤化物、VI 族卤化物、HI、HBr、HCl 以及它们的组合。在另一个实施方案中，与基体

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活性碳结合时的促进剂基本上是蒸汽形式的。在另一个实施方案中，促进剂在与基体活性碳反应之前与有机溶剂结合。在另一个实施方案中，促进剂和任选的次级组分基本上同时与基体活性碳结合。另一个实施方案中还包括加入选自 S、Se、H_2S、SO_2、H_2Se、SeO_2、CS_2、P_2S_5 以及它们的组合的汞稳定剂。另一个实施方案中还包括加入任选的碱性组分。

在一个实施方案中提供了一种方法，其包括：提供粒状的活性碳；使活性碳与选自卤素、卤化物以及它们的组合的促进剂反应，使反应产物包含能从气流中有效清除汞的促进的碳吸附剂。在进一步的实施方案中，相对于每 100 克活性碳，反应产物中包含约 1 至约 30 克促进剂。在另一个实施方案中，反应产物具有的平均粒度分布大于欲清除汞的气流中夹带的灰分粒子的平均尺寸，从而使得可以通过物理方法将反应产物从夹带的灰分粒子中基本移出。在另一个实施方案中，反应产物具有大于约 40 微米的质均粒径。

在另一个实施方案中，促进剂选自分子态卤素、氢卤化物、V 族卤化物、VI 族卤化物以及它们的组合。在另一个实施方案中，促进剂在接触活性碳时是在气相中。在另一个实施方案中，促进剂在接触活性碳时在有机溶剂中。

在另一个实施方案中，促进剂选自 Br_2、V 族溴化物、VI 族溴化物以及它们的组合。

在另一个实施方案中，所述方法还包括使粒状的活性碳与任选的次级组分进行反应，所述次级组分包括卤素或氢卤化物，从而使吸附剂的活性和汞容量得以增加。在另一个实施方案中，促进剂和任选的次级组分同时与活性碳接触。在另一个实施方案中，所述方法还包括加入选自 S、Se、H_2S、SO_2、H_2Se、SeO_2、CS_2、P_2S_5 以及它们的组合的汞稳定剂。在一个实施方案中提供了一种以较低的清除速率控制烟

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道气中汞的方法，所述方法对吸附剂的要求充分的低。通过吸附剂活性的增强，每克吸附剂的汞清除量也增加，因此，通过降低对吸附剂的要求可以降低投资和操作成本。

在一个实施方案中提供了一种减少烟道气中的汞的方法，所述方法包括提供吸附剂、将吸附剂注入到包含汞的烟道气流内、在吸附剂上收集超过 70 wt%的烟道气中的汞以产生清洁的烟道气以及从清洁的烟道气中基本回收吸附剂。在期望汞的清除低于 70 wt%的实施方案中，优选可以通过使用低于采用标准(非增强的)碳时所需碳量的一半的碳来达到所需的清除 。在进一步的实施方案中，所述方法还包括监测清洁的烟道气中的汞含量、使回收的吸附剂再生及应用监测的清洁的烟道气中的汞含量来控制吸附剂的注入速率。在另一个实施方案中，通过使活性碳与促进剂在气动的运输管线内反应来动态地制备注入的吸附剂，反应产物从该管线中注入到包含汞的烟道气流内。

在另一个实施方案中，促进剂选自分子态卤素、卤化物以及它们的组合。在另一个实施方案中，促进剂在气相中或以蒸汽形式进行反应。在另一个实施方案中，相对于每 100 克的活性碳加入约 1 至约 30 克促进剂。

在另一个实施方案中，通过在气动的运输管线内使活性碳、促进剂和任选的次级组分进行反应来动态地制备注入的吸附剂，从而增加吸附剂的活性和容量，反应产物从该管线中注入到包含汞的烟道气流内。

在另一个实施方案中，任选的次级组分选自碘、氢卤化物、V 族卤化物、VI 族卤化物以及它们的组合。在另一个实施方案中，任选的次级组分的加入量为促进剂含量的约 1 至约 15 wt%。在另一个实施方案中，所述方法还包括在吸附剂中加入选自 S、Se、H_2S、SO_2、H_2Se、SeO_2、CS_2、P_2S_5 以及它们的组合的汞稳定剂。

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在一个实施方案中，所述方法还包括共注入任选的包括但不限于碱和碱土组分的碱性物质，从而通过捕集被氧化的汞和/或捕集可能会降低吸附剂容量的气态组分来改善汞捕集效率。在另一个实施方案中，任选的碱性物质优选可以包括氧化钙、碳酸钠等等，如在本领域中已知的那些。

在另一个实施方案中，所述方法还包括应用监测的清洁的烟道气中的汞含量来控制吸附剂的组成。在另一个实施方案中，通过使活性碳与促进剂在气动的运输管线内反应来动态地制备注入的吸附剂，反应产物从该管线中注入到包含汞的烟道气流内，其中促进剂选自分子态卤素、卤化物以及它们的组合，其中促进剂在气相中反应或以蒸汽形式进行反应，其中相对于每 100 克的活性碳加入约 1 至约 30 克促进剂，其中至少部分地基于清洁的烟道气中的监测汞含量，通过数字计算机来确定促进剂的加入速率和吸附剂注入速率。

在一个实施方案中，提供了减少包含汞及灰分的气流中的汞含量的方法，其中将质均尺寸大于 40 μm 的微粒活性碳吸附剂注入到气流中，通过吸附剂粒子从气体中清除汞，基于尺寸大小使吸附剂粒子与灰分粒子分离，将吸附剂粒子再注入到气流中。在另一个实施方案中，使包含汞的吸附剂粒子再生，从而清除一些或基本上所有的汞。在另一个实施方案中，将碱性组分共注入到气流中。在另一个实施方案中，吸附剂可以还包含促进剂。促进剂优选可以包含卤化物、卤素，或包含它们二者。

如以下将更详细描述的那样，本发明从而提供了优于以前已知技术的若干优点，包括对流出气体显著更有效及经济性的汞吸附剂，可以有利地适用于燃烧设备及气化系统的气流处理。

为了可以更好地理解随后对本发明的详细描述，前述内容已经相

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当广泛地概括了本发明的特点和技术优势。以下将描述本发明另外的特点及优势，其构成本发明的权利要求的主题。本领域技术人员应该理解的是，可以很容易地利用所公开的概念和具体实施方案为基础，修改或设计出其它的结构以实现与本发明相同的目的。本领域技术人员还应该意识到，这种等价的构造并不偏离随附的权利要求中限定的本发明的实质和范围。

附图说明

本发明优选实施方案的更详细描述可参考所给出的附图。

图 1 示意性地例示根据本发明的促进的碳吸附剂的制备方法。

图 2 例示了提出的导致汞氧化及捕集的化学反应的机理模型。

图 3 示意性地例示根据本发明的促进的碳吸附剂的制备以及降低气化系统的产物气和/或烟道气中的烟道气汞的方法，包括促进的碳吸附剂的动态制备。

图 4 为例示 5 wt/wt %溴化 NORIT Darco FGD 吸附剂(37 mg+113 mg 砂) 在低-HCl (1 ppm) 合成烟道气中的穿透曲线的图示。

图 5 为例示非卤化 NORIT Darco FGD 吸附剂(37 mg+113 mg 砂) 在低-HCl (1 ppm) 合成烟道气中的穿透曲线的图示。

图 6 为例示中试规模汞清除结果的条形图表，包括大尺寸吸附剂的结果。

图 7 为例示对于 ESP 和织物过滤器而言的吸附剂尺寸和注入速率对汞清除的影响的图示。

图 8 为例示具有惰性砂的溴化 NORIT Darco FGD 吸附剂的穿透曲线的图示。

图 9 为例示具有共注入碱性物质的溴化 NORIT Darco FGD 吸附剂的穿透曲线的图示。

图 10 为具有和不具有共注入的碱性物质时的汞清除对碳注入速率的图示。

优选实施方案的详细描述

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此处将详细描述本发明具体的优选实施方案，须理解的是，应该认为此处的公开内容为本发明原理的例证，并不意味着将本发明限制在此处例示和描述的内容上。本发明容许不同形式或次序的优选实施方案，不应该认为限制在具体表述的方法或本文所包含的组分上。特别而言，本发明各种优选的实施方案提供了本发明方法、成分以及它们的用途的许多不同构造和应用。

本发明提供利用异常活性的卤素/卤化物促进的碳吸附剂来捕集污染物的成本有效的方法，所述吸附剂使用溴化物(或其它卤素/卤化物)处理的碳，其在若干秒或更短的接触时间内通过汞-吸附剂表面反应来捕集汞。与常规的活性碳吸附剂不同，该吸附剂不需要在气流中的原位活化以达到较高的活性（没有诱导期）。吸附剂对污染物的活性大大增加，可以恢复吸附容量， 可以再生、再循环和/或重新使用促进的吸附剂。

除了对于燃烧系统的烟道气之外，此处讨论的处理的碳、处理技术以及任选的添加剂对于产物气或排出气或者气化系统、合成气发生炉的气体以及其它含汞气流中的汞控制也具有适用性。因此，应该理解的是，本说明书中全文中使用的术语燃烧系统和烟道气可同样地应用于气化系统以及合成气或燃料气，如本领域的技术人员将会理解的那样。

现在参考图 1，显示的是例示本发明方法的一些优选实施方案的方框流程图，用来制备用于从气化系统流的产物气和/或烟道气中捕集汞的促进的吸附剂。在一个由路径 10-20 例示的优选实施方案中，方框 10 例示的是提供基体活性碳，方框 20 例示的是加入与碳反应的卤素或卤化物促进剂，以生成产物促进的碳吸附剂。在以例如蒸汽的形式加入卤素或卤化物的实施方案中，可不需要有另外的步骤。在以例如溶剂的形式加入卤素或卤化物的实施方案中，则可以期望采用如方框 20A 例示的溶剂清除。

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仍然参考图 1，路径 10-20-30 例示了本发明方法的另一个优选的实施方案，包括如方框 10 表示的提供基体活性碳、方框 20 例示的加入与碳反应的卤素或卤化物促进剂以及方框 30 例示的加入与方框 20 的产物反应的次级组分，以生成产物促进的碳吸附剂。在卤素或卤化物促进剂以及次级组分均以例如蒸汽形式加入的实施方案中，可以无需另外的步骤。在以例如溶剂的形式加入卤素或卤化物促进剂和/或次级组分的实施方案中，则可以期望采用如方框 30A 例示的溶剂清除。

仍然参考图 1，路径 10-40 例示了本发明方法的另一个优选的实施方案，其包括如方框 10 表示的提供基体活性碳、方框 40 例示的将卤素或卤化物促进剂以及次级组分一道加入至与之反应的活性碳中，生成产物促进的碳吸附剂。如上所述，在采用蒸汽加入至活性碳的实施方案中，可以无需另外的步骤。在以溶剂形式加入一种或多种组分的实施方案中，则可以需要提供如方框 40A 例示的溶剂清除步骤。

仍然参考图 1，其还例示了优选实施方案，其中如方框 50 所例示，采用如上所述制备的产物促进的碳吸附剂来处理烟道气流。

本领域技术人员可以理解的是，在一些优选的实施方案中，所提供的活性碳优选可以为若干类型中的任何种类。例如，活性碳可以包括粉状活性碳、粒状活性碳、碳黑、碳纤维、碳蜂窝或板状结构碳、气凝胶碳薄膜、热解炭、源于产物促进的碳吸附剂的再生活性碳或本领域中已知的其它类型。

本领域技术人员可以理解的是，在一些优选的实施方案中，所提供的活性碳优选可以为若干类型中的任何种类。例如，活性碳可以包括粉状活性碳、粒状活性碳、碳黑、碳纤维、碳蜂窝或板状结构碳、气凝胶碳薄膜、热解炭、质均粒度大于欲处理烟道气流中的飞灰的活性碳或再生活性碳。

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本领域技术人员可以理解的是，在一些优选的实施方案中，所提供的活性碳优选可以为若干类型中的任何种类。例如，活性碳可以包括粉状活性碳、粒状活性碳、碳黑、碳纤维、碳蜂窝或板状结构碳、气凝胶碳薄膜、热解炭、活性碳或再生活性碳，所述活性碳或再生活性碳的质均粒径优选大于 40 微米，更优选大于 60 微米，或者粒度分布比欲处理的烟道气流中的飞灰或夹带灰分更大，从而使得可以通过物理方法来分离活性碳和灰分。

作为例示而非限制，在一些优选的实施方案中，加入到基体活性碳中并与之反应的卤素或卤化物促进剂优选可以包括：蒸汽或气体形式的分子态卤素；在有机溶剂中的分子态卤素；蒸汽、液体或溶液形式(但不在含水溶剂中)的 V 族或 VI 族卤化物，例如分别是 PBr_3 或 SCl_2。

还提供了这样的实施方案，其中有机溶剂优选可以包括氯代烃如二氯甲烷，例如包括石油醚、轻石油、戊烷、已烷、甲苯和苯的烃溶剂，二硫化碳，废弃溶剂，醚，再循环溶剂，超临界溶剂如超临界 CO_2，水(但不是在 V 族或 VI 族卤化物的情况下)，以及对于本领域的技术人员显而易见的其它溶剂。

现在参考图 2，其例示了由科学性的论据导出的理论，用以解释促进性化合物的性质。例如，如图 2 所例示，溴化氢与活性碳的不饱和结构发生反应。仅作为例示而言，这可以是位于碳的石墨烯(graphene)片层结构的边缘上的碳烯物质。分子态溴或溴化合物的反应可形成类似的结构，带有对汞的氧化具有活性的正碳，随后被吸附剂所捕集。

现在已经发现，与碳形成新的溴化合物增大了它们对于汞和其它污染物的活性。另外，所得到的溴化合物对于促进汞的氧化具有独特的适用性。氧化的有效性显然是源于卤化物对氧化期间在汞上逐渐发展的正电荷产生的促进效果，这在化学领域中被认为是特定催化效应。

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因此，当汞的电子被拉向正碳时，卤化物阴离子的电子则从另一侧推进，使在汞上逐渐发展的正电荷稳定化，降低氧化过程所需的能量。溴化物是特别有活性的，这是由于其离子的外层 4p 轨道上的可高度极化的电子的缘故。因此，在碳上加入 HBr 或 Br_2 形成类似的碳溴化物，其中正碳在溴离子的协助下将汞氧化。

现在参考图 3，提供了汞控制系统 100 的示意流程图，依照本发明优选的实施方案，其包括制备促进的碳吸附剂和降低烟道气中的汞。提供了基体活性碳储槽 110、任选的卤素/卤化物促进剂储槽 120、任选的次级组分储槽 130 和任选的碱性组分储槽 180，它们的每一个都带有各自相应的流动控制装置 201、202、203 和 208/209。对于任选的碱性组分储槽 180，可以独立地使用任选的流动控制装置 208 和 209，或一起使用它们，或者不使用它们。

储槽 110、120、130 和 180 通过它们各自的流动控制装置经由相关的管道连接至运输管线 115。任选的碱性组分储槽 180 也可以通过各自的流动控制装置经由相关的管道连接至运输管线 118。通过气体源 170 将空气、氮或其它运输气体源提供至运输管线 115，使之夹带从储槽 110、120、130 和 180 中排出的物料，并经由注入点 116 将这种物料注入到污染的烟道气流 15 之内。可以通过气体源 171 将空气、氮或其它运输气体源提供至运输管线 118，使之夹带从储槽 180 中排出的物料，并经由注入点 119 将这种物料注入到烟道气流 15 之内。根据需要，气体源 170 和 171 可以相同或不同。或者，可以通过气体源 170 将运输气体提供至运输管线 115 和 118 两者(从源 170 至管线 118 的连接未示出)。虽然在图 3 中显示的气体源 170 和 171 为压气机或鼓风机，但本领域的技术人员将会意识到，本领域中已知的任何运输能量源都是可以接受的。

为清楚起见，在图 3 中显示了单一注入点 116 或 119，本领域的技术人员可以理解的是，多重注入点也在本发明的范围之内。光密度

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测量装置 204 连接至运输管线 115 和/或 118，从而提供代表运输管线 115 和/或 118 内部的光密度的信号，其为时间的函数。

注入点 116 和 119 的下游提供了微粒分离器 140。以说明而非限制性的方式，微粒分离器 140 可以包括一种或多种织物过滤器、一种或多种静电沉降器(以下称“ESP”)或本领域中已知的其它微粒清除装置。还应该指出的是，可以存在顺序配置或并列配置的一个以上微粒分离器 140，当存在并列配置、顺序配置或它们的组合配置时，注入点 116 和 119 可以在 140 的上游和/或下游的位置。微粒分离器 140 至少产生主导性的气态(“清洁”) 流 142 和包含分离的固体物质的流 141。吸附剂/灰分分离器 150 将流 141 分离成大体为灰分的流 152 和大体为吸附剂的流 151。然后优选可以使流 151 通过任选的吸附剂再生器 160，其产生再生吸附剂流 161 和废物流 162。

在废气流 35 中提供任选的汞连续排放监测器(以下称“CEM”) 205，用以提供代表废气流 35 中汞浓度的电信号，其为时间的函数。任选的汞 CEM205 与流动控制器 201、202、203、208 和 209 经由任选的线路 207(或无线地)电连接至任选的数字计算机(或控制器) 206，其接收和处理信号，并优选控制促进的碳吸附剂的制备及其向污染的烟道气流 15 内的注入。

在操作中，将促进的碳吸附剂和/或任选的碱性组分注入到污染的烟道气流 15 内。在注入的物质与污染的烟道气流 15 接触以后，注入的物质使汞的浓度降低，将污染的烟道气转变为汞降低的烟道气 25。通过分离器 140 从烟道气 25 中清除注入的物质，分别地将其处理掉或通过任选的分离器 150 进行进一步的分离，再处理掉或通过任选的再生器 160 进行再生。然后，汞降低的“清洁”烟道气流 142 中采用任选的 CEM205 监测汞含量，其向任选的计算机/控制器 206 提供相应的信号。然后，依照本领域中熟知的控制算法，使来自 206 中的逻辑及最优化信号调节流动控制器 201、202、203、208、209，从而保持废气

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流 35 中汞浓度在期望的限度以内。流动控制器 201、202、203、208、209 也可以是手动调节的或某些其它的自动化装置，从而依照本领域中熟知的控制算法以保持废气流 35 中汞浓度在期望的限度以内。

仍然参考图 3，例示了依照本发明的促进的碳吸附剂和/或碱性组分的制备及注入的若干优选的实施方案。流 111 提供从储槽 110 的基体活性碳的引入，这通过流动控制器 201 手动地或者在计算机 206 的指令下进行计量。根据任何所提供的若干方法，可以使卤素/卤化物和基体活性碳结合并与之进行反应。卤素/卤化物优选可以经由管线 121 直接结合到运输管线 115 内，在其内于注入点 116 之前和基体活性碳接触并与之反应。依照本发明，该选择为本文所指的促进的碳吸附剂的“动态”制备的一种形式。进一步地，卤素/卤化物在进入运输管线 115 之前可以经由管线 121b 与基体活性碳结合。更进一步地，卤素/卤化物可以通过经由管线 121c 引入到储槽 110 内而和基体活性碳接触并与之反应。当例如储槽 110 包括基体活性碳的沸腾床或流化床时，卤素/卤化物在其中以气体的形式或作为蒸汽流动，该选择是优选采用的。当然，如前面讨论到的那样，卤素/卤化物也可以优选以液体的形式或在溶剂中与基体活性碳接触，然后，如果需要的话，如参考图 1 所讨论的实施方案所提到的那样，可以提供溶剂清除(图 3 中未示出)。

类似地，任选的次级组分可以经由管线 131、或任选地如上面所描述的卤素/卤化物那样经由管线 131b 和 131c 而直接在运输管线 115 中接触与反应。

类似地，180 中任选的碱性组分既可以直接加入到运输管线 115 中，或者也可以通过运输管线 118 单独地注入，在下游结合到烟道气 15 中，与基体活性碳、促进的碳或任选的次级组分产生协同效应。能够相对于基体活性碳、促进的碳或任选的次级组分而原位地改变任选的碱性组分的量，这是克服位置特异性操作及烟道气条件并优化之的一个关键性特征。

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在一些其中组分之间的接触及反应是在液体或溶剂相中进行的优选实施方案中，可以提供对这种液体和/或浆料混合物的搅拌。在其它的实施方案中，优选可以溶液的形式将卤素/卤化物促进剂和任选的次级组分喷射到基体活性碳之内或之上。在一些这样的实施方案中，然后可以提供干燥、过滤、离心、沉降、倾析或本领域中已知的其它溶剂清除方法。

在其中卤素/卤化物促进剂为气体或蒸汽形式的实施方案中，可视情况将其在空气、氮或其它气体中进行稀释。可以使例如气态 HBr 或 Br_2 的卤化物/卤素气体通过粒状或纤维状活性碳的沸腾床或流化床，经由气体夹带将如此生成的促进的碳吸附剂从床的顶部移走，用于注入。

在一些实施方案中，次级组分优选可以包括碘或其它卤素，氢卤化物，包括但不限于 HI、HBr、HCl，V 族或 VI 族元素与分子态卤素，例如 SCl_2 及其它。在一些优选的实施方案中，相对于每 100 g 基体活性碳，促进的碳吸附剂可以包含约 1 至约 30 g 的卤素/卤化物。在一些优选的实施方案中，促进的碳吸附剂可以包含次级组分，其浓度为卤素/卤化物组分浓度的约 1 至约 15 wt%。

还在其它的实施方案中，可以将产物促进的碳吸附剂施加到基材上。在其它的实施方案中，为了达到降低汞的目的，可以使这样制备的基材与污染的烟道气或气化系统的产物气流接触。这样的基材可以是单片的(monolithic)、旋转的，或者以本领域技术人员已知的任何方式暴露于气流中。

在一些实施方案中，提供了这样的方法，其中汞稳定剂加入到促进的碳吸附剂中，从而产生双功能吸附剂。可以在卤素/卤化物的加入及反应之前或之后顺序地加入这种稳定剂。在一些优选的实施方案中，卤素/卤化物优选包括 Br 或 HBr，汞-稳定剂可以包括 S、Se、H_2S、SO_2、

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H_2Se、SeO_2、CS_2、P_2S_5以及它们的组合。

汞的捕集中的卤素

在使用卤素来处理烟道气的方法学上已经产生一些问题，这是由于它们对其它气体及金属具有的活性，导致腐蚀及健康方面的问题。“卤素”定义为一系列的非常活跃的元素，包括周期表中的 VIIA 族(通篇使用 CAS 命名；VIIA 族(CAS)对应于 VIIB 族(IUPAC))。分子态元素形式的卤素，包括 F_2、Cl_2、Br_2和 I_2，其与热烟道气组分的反应后，几乎没有剩余可与元素汞反应。原子态元素的卤素形式包括氟、氯、溴和碘原子，其对汞的反应活性要大出约一百万倍，但原子态形式的浓度通常是极低的。在大部分的电业煤炭燃烧设施中，其浓度通常不足以使显著量的汞被氧化。

此处采用的术语“卤化物”定义为由卤素与另一种元素或基团反应形成的化合物。一般地，卤化物比起分子态卤素来讲活性要低得多，具有较低的化学势。卤化物被认为是不能单独氧化其它化合物的还原态形式。因此在传统的观点当中，卤化物-盐-处理的活性碳不能有效地将元素态的汞氧化及捕集元素态的汞。

卤素促进的吸附剂特性

这里描述的吸附剂对于汞的氧化具有非常高的初始活性，因此使用非常少的量就可以得到非常高的捕集效率，从而降低操作成本，减少废物处理的问题。此外，对采用本发明技术生成的吸附剂进行再生及重新使用可以获得进一步的处理减少。汞与本发明的促进的碳吸附剂在烟道气导管中顺利地相互作用以及随后汞在吸附剂和灰分上的收集所需的时间间隔非常短，不到几秒钟。很显然，这样的收集时间需要吸附剂对汞既具有高容量又具有高活性。可以非常细的粉末形式利用促进的碳吸附剂，从而使传质限制最小化。但此外，活性应该非常之高，从而捕集细粒子所遇到的所有的汞。另外，采用这些增强技术可以容许得到对更大吸附剂粒子而言的有效捕集，而更大的吸附剂粒

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子使得吸附剂能从灰分中分离，从而能够进行随后的再生以及灰分的利用。本发明的一个特点在于制备含有在碳结构上形成的卤化物的吸附剂的方法，其提供与汞污染的气流初始接触时具有高度活性的吸附剂，这使得能非常有效地捕集汞。

看起来本发明的吸附剂用活性碳(边缘位置)例如从溶液中与分子态溴化学地结合。X 射线光电子能谱法已经确定，溴、氯、HBr 或 HCl 的加入在碳结构中形成了化学的化合物。因此，由卤素和活性碳生成的吸附剂不再是分子态卤素的形式，而是一种新的化学改性的碳(或卤化碳)结构。除卤化物离子外，在低活性的碘中可能不出现这种现象，其中 I_2 分子络合物可能存在于碳基面上。在溴的情况下，改性的阳离子碳具有使汞氧化的高化学势。因此而给出了溴处理的碳对汞的活性的全新模型。优选可以通过加入溴、溴化氢或如本文中描述的溴与其它元素的组合来产生活性的碳形式。卤素处理产生具有更高活性的碳，因为卤化物(特别是溴化物和碘化物)阴离子通过使在氧化的过渡态中汞上逐渐发展的正电荷稳定化而有效地促进氧化。基于该模型而产生若干创新、价廉、活性增强的特点。

任选的次级组分

已经证实，除了溴以外任选的次级组分的加入导致吸附剂活性和容量的改善，通常超过了未处理的碳和溴化的碳。次级化合物或者包括次级卤素，或者包括次级卤素衍生的化合物，例如 HI。因此，除了存在具有活性的碳形式外，次级组分产生 Lewis 碱，其具有更强的能力以使在汞上逐渐发展的正电荷稳定化。因此，次级组分为具有更大程度极化电子(4p 和 5p)的元素。

任选的碱性组分

已经证实，在基体活性碳或促进的活性碳中加入任选的碱性组分可以导致汞捕集的改善，通常要超过未处理的碳和促进的碳。测试数据表明，烟道气污染物、烟道气成分(SO_2、NO_x、HCl 等)、操作温度、

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汞的形式以及汞的浓度可以影响到碱加入的有效性。这就暗示需要能够原位地调节及定制碱与活性碳的比率，从而能克服给定的原位条件并优化之。

可以如下地解释当共注入两种材料时得到的协同。首先，测试表明，活性碳(以下称作“AC”)上的结合位置可以被氯物质、硫物质(即硫酸盐)和其它的烟道气污染物(砷酸盐、硒酸盐等)所消耗。加入的任选碱材料将与这些物质/污染物相互作用及反应，从而使它们对于 AC 汞的结合位置的消耗最小化。其次，测试还表明，即使结合位置完全被消耗，标准 AC 也将继续进行汞的氧化。然后该被氧化的汞可以与碱性物质反应，并随后被粒子控制装置所捕集。因此，任选的碱性组分的加入起到保护汞结合位置及捕集被氧化的汞的作用，从而导致在更低成本下的汞降低的改善。与活性碳相比，碱成本通常要低得多(差了~一个数量级)，因此可以更多地使用，而总体成本仍然较低。

“动态的”吸附剂制备

此外，已经证实可以容易“动态地”生成卤素促进的碳吸附剂。这可以通过以下来实现，例如，使卤素和任选次级组分的任意组合的蒸汽动态地与非常细的碳粒子接触。粒子可以被分散在运输空气(或其它气体)流中，该气流也将卤素/卤化物促进的碳吸附剂粒子输送至烟道气导管或其它污染的气流中，然后汞从那里被清除。对于该接触没有特别的温度要求。该技术的实施显然非常简单，并在很大程度上导致将该技术用于汞捕集的设备成本的节省。

原位制备的优点

在原位置动态地制备卤素/卤化物促进的碳吸附剂具有某些优点。例如，处理系统可以在最终使用位置上与碳注入系统结合。采用此技术时，卤素/卤化物被引入到运输管线(或吸附剂存储及注入系统的其它部分)中的碳-空气(或其它气体)混合物中。这提供了如下优于现有的离位处理吸附剂的常规概念的益处：

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●排除了处理设施的投资设备成本

●排除了操作处理设施的的成本

●没有关于将碳和添加剂运输到处理设施的成本

●本发明方法使用现有的硬件及操作步骤

●本发明技术确保吸附剂始终是新的，因此更具有活性

●没有引入新的处理方面的问题

●没有从处理系统中清除碳的成本

●本发明方法可以容许添加剂-吸附剂比的快速原位定制，从而可以适应例如当改变燃料或减少负载时可能需要的烟道气变化的要求，因此可进一步优化经济性

●本发明技术使需处理的用过的吸附剂量减少

考虑到前述内容以及其它特点，根据本发明，提供了以下实施方案，包括制备和再生卤素/卤化物促进的碳吸附剂的方法，通过将卤素(例如溴)加入到碳结构中增强其汞捕集的活性。

吸附剂注入位置

一些优选实施方案设想以粉末形式使用卤素促进的吸附剂，其在清除灰分微粒之前或之后注入到烟道气流中。本发明其它实施方案的卤素促进的碳吸附剂的组成包括通过加入 Br_2 或 HBr 和任选的次级组分而制备的粉状改性活性碳。其它实施方案容许与基体活性碳协同地加入任选的碱性组分、和/或使用卤素基吸附剂，以及本专利中提供的吸附剂技术的任意其它的组合。或者，实施方案包括这样的方法，其中吸附剂在由含有一种或多种上列组成的粒子或纤维组成的移动接触器上。

吸附剂再生

可以很容易地使任何上述实施方案的卤素/卤化物促进的碳吸附剂再生；优选清除烟道气中的毒害性污染物，加入便宜的促进剂，从而恢复汞吸附活性。该促进碳本身的活性的方法与在吸附剂中加入试剂(例如过氧化物、金、三碘化物等)的早期更昂贵的常规方法形成对比。

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本发明的卤素/卤化物促进的碳吸附剂是用溴和/或任选组分处理的，是非腐蚀性的。吸附剂再生技术的详细实例描述于标题为“PROCESS FOR REGENERATING A SPENT SORBENT”的共同未决、共有的PCT专利申请No. PCT/US04/12828中，现将其全文引入以供参考。

吸附剂注入控制方案

本发明另一优点涉及使用反馈系统来更有效地利用本发明的某些方面。在可能和期望的情况下，本发明的汞控制技术优选可以利用汞排放的连续测量作为反馈，协助控制吸附剂注入速率。可以这样地达到吸附剂和任选组分水平的更严格控制，这将保证汞清除的条件符合最低的物质要求，从而使相关成本最小化。在一个实施方案中，在注入位置的下游，优选在烟道的废气中连续测定汞排放。

促进的碳吸附剂

卤素和酸性物质与活性碳吸附剂上的碱性结合位置的反应产生了将汞氧化的位置。诸如硼、锡、砷、镓、Sb、Pb、Bi、Cd、Ag、Cu、Zn的其它金属离子或其它污染物也将与碳上产生的氧化位置进行反应。

根据我们的模型，将溴试剂中的溴或卤化氢酸中的质子加入到碳边缘结构上的碱性碳烯位置上，以形成碳正离子，其接受中性汞原子中的电子，形成结合到吸附剂表面上的氧化了的汞物质。活性位置还可以在碳的活性位置上产生活性的溴基或碳基。因此活性碳用于使溴稳定化，但能提供高度活性的含溴试剂，该试剂可以使汞氧化并促进其在活性碳上的捕集。预计包含溴的吸附剂比相应的包含氯的吸附剂活性大，且比包含碘的吸附剂便宜得多。

实施例

为了更清楚地例示本发明，下面给出若干实施例。这些实施例是作为例示目的的，本文中给出的实施例不应该理解或推断为对本发明

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的限制。

实施例 1-制备及测试卤化的碳(及比较例)

气相卤化

将细的粉末活性碳(例如 NORIT Darco FGD，NORIT Americas，Inc.，Marshall，Tex.(USA)，尽管如本领域技术人员可认识到的那样，也可适用其它的)置于带有侧叶片的旋转式塑料桶(5 ft^3 (0.14 m^3) 水泥搅拌器)中，其上安装有紧密的塑料盖以防止在制备期间细粉末的损失。在一个单独的容器中，气相溴的制备是通过使氮流流过被暖热至约 40℃-50℃的已称重量的液体溴来实现的。溴的蒸汽压力使得产生的暗红色气体流出发生器。气态溴发生器的出口经由一个 1/4 英寸(0.64 cm)的塑料胶管连接至固定的金属管，所述金属管通过法兰插入到塑料盖的中心并穿入到桶的中心内部。法兰不是空气密闭的，当溴被传递至翻转的碳上之后，过剩的氮被释出。如此，溴气流不断地流入到旋转桶的内部，在那里与翻转的碳接触。然后操作该单元，直到期望数量的溴与碳结合。通常是，0.4 至 1 kg 的溴对 20 kg 的碳(2-5 wt. %)。当反应完毕时，称量碳的重量。处理过的碳是无嗅的，不会产生皮肤刺激性，因为溴已经完全与碳反应而生成了溴化的碳。

XPS 光谱表明，溴化的碳包含与碳键合的(有机)共价溴化物以及阴离子溴化物。产物中包含的湿度与初始存在于活性碳中的相同(5-17 wt %)，但无需进一步的干燥即可使用。在更高的温度下(>150℃)湿度被去除，直到非常高的温度时才会释放出溴。

汞的氧化及捕集效率的实验室规模测试

使用基于以上描述的实验室规模的装置和步骤测试若干促进的活性碳吸附剂的初始活性及容量， 所述吸附剂使用粉状碳，包括由多种碳制备的含溴活性碳，包括商业上可得到的吸附剂、气凝胶膜吸附剂和初始前体碳用于比较。

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文献 Dunham，G.E.；Miller，S. J. Chang，R.；Bergman，P. *Environmental Progress* 1998，17，203 提供了装置及其操作的详细描述，在此全文引入以供参考。用细粉(-400 目)状吸附剂(37 mg)与 113 mg 砂混合，装填在石英过滤器(2.5 英寸(6.35 cm))中，进行烟道气组合物中的实验室规模汞吸附剂测试。在模拟的烟道气流(30 SCFH (标准立方英尺/小时)或 0.79 NCMH (标准立方米每小时))下将装填的过滤器和支架于炉中(125℃)加热，所述模拟的烟道气流包含如下成分：O_2 (6%)、CO_2 (12%)、SO_2 (600 ppm)、NO (120 ppm) NO_2 (6 ppm)、HCl (1 ppm)、Hg^0(11 μ g/m^3)、H_2O (15%)和 N_2 (余量)。通过置于双夹套玻璃冷凝器中并加热至所期望温度的标准渗透管源提供元素汞。使用连续汞排放监测器(Sir Galahad 汞 CEM，制造商 P.S. Analytical Deerfield Beach FL USA)确定气流中的汞浓度，并使用 $SnCl_2$ 池将被氧化的物质转变为元素态，从而可以获得对于从吸附剂床的流入及流出浓度的元素态及被氧化的汞的浓度数据。根据采用的流速校准汞浓度。对用过的吸附剂进行汞的分析以确定质量平衡。

现在参考图 4，给出作为流入汞百分数的流出汞浓度数据对时间的图示。卤化的吸附剂所得到的曲线(穿透曲线)显示流出物中最初有 0%-1%的 Hg(99+% 捕集)，仅在 30-60 分钟(穿透点)之后有所增加，这取决于吸附剂情况。图 4 例示的是对于 5 wt/wt %溴化的 NORIT Darco FGD 吸附剂(37 mg+113 mg 砂)与包含 1 ppm HCl 的合成烟道气的穿透曲线。流出物中总 Hg(实心圆)和元素态 Hg(实心方框)给出为入口 Hg 的百分数。“EOT”表示测试的终点(后面显示的数据点用于校准核对)。

图 5 给出的是依照合成烟道气中的 HCl 浓度情况，通常起始为 5%-50%入口汞时相应的非卤化吸附剂的比较穿透曲线，由此表明了非卤化吸附剂具有相当低的汞氧化及捕集活性。无论是卤化或非卤化的吸附剂，在穿透以后，流出物中大部分的汞为被氧化的汞。

实施例 2 -流态化碳的气相卤化

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通过氮流使由玻璃棉塞支撑在垂直管中的活性碳床流态化。流化床管的顶部与用于吹出管顶部的碳细粒的捕集器连接。如实施例 1 中描述的溴气体发生器与流化碳床相连，使期望数量的气态溴流到床内。然后将捕集器中的内含物与床内的物质混合并称重。得到的溴化的碳表现出与实施例 1 中溴化的碳类似的性能。

实施例 3 -液相(水)卤化

将 50 g 溴小心地加入到 1 升冷水中，制备溴的 5%水溶液。将一 kg 的活性碳加入到大金属罐里的溴溶液中。在加入的期间用大桨叶片对得到的浆料搅拌，经短时间后，直到红色消失表明所有的溴已经与碳反应。然后用布氏漏斗真空过滤浆料。将过滤器中收集到的潮湿碳在 110℃炉中干燥若干小时至恒重。如在实施例 1 中那样，碳中还仍然留有一定湿度。然后使干燥的碳在旋转桶中翻转，利用金属片破碎及疏解(fluff)碳。

实施例 4 -任选次级卤化物组分的加入

通过类似于根据实施例 3 所描述的溶液相溴化生成溴化的碳。然而在过滤之前，将氢碘酸(HI)溶液加入到浆料中，其量等于溴量的 10%。搅拌浆料以完成反应，然后如实施例 3 中描述的那样进行过滤及干燥。

实施例 5-液相磷卤化

在大金属罐中搅拌三溴化磷(500 g)的轻石油(10 升)溶液，加入 10 kg 的活性碳。用大桨叶片在环境温度下对所得到的浆料进行搅拌以完成反应。以若干批次在大的布氏漏斗中真空过滤浆料。湿滤饼在 110℃炉中干燥至恒重。干燥的产物如在实施例 3 中所描述的那样在旋转桶中疏解。

实施例 6-更大粒子碳的制备及其上的吸附

燃烧次烟煤炭，用中试规模的燃烧器进行测试，通过注入大于正常尺寸的处理的活性碳来评估汞的控制。标准 AC 吸附剂通常为细尺寸

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的，平均粒径小于 20 微米，这也是煤粉燃烧产生的飞灰的典型尺寸。因此，由于标准 AC 与飞灰的尺寸类似，两者的分离是困难的。通常不考虑注入更大尺寸的 AC，因为吸附剂的有效性随尺寸而降低。在使注入的碳再循环的方案中，从飞灰中分离出碳。基于尺寸分级的分离需要处理的更大粒子的吸附剂。为了验证这种观点，开发、制备和测试了处理的更大尺寸的(>60 μ m)吸附剂。

处理-气相卤化

研磨粒状活性碳(Calgon F400)并用常规网筛进行筛分。收集-170至+240 的网目尺寸部份(相当于约 60 至约 88 微米)并置于如上面实施例 1 中描述的旋转容器中。也如在实施例 1 中描述的那样，在单独的容器中，气相溴的产生是通过使氮流在被暖热至约 40℃-50℃的已称重量的液体溴之上流过来实现的，该气态溴发生器的出口经由一个 1/4 英寸(6.35 mm)的塑料胶管连接至固定的金属管，所述金属管通过法兰插入到盖的中心并穿入到旋转容器的中心内部。操作该单元，直到期望量的溴与碳结合，在这里是 0.05 kg 的溴与 1 kg 的碳(5 wt.%)。当反应完毕时，称量碳的重量。如上面已经描述的那样，处理的碳是无嗅的。

PTC 装置

中试规模燃烧器即已知的“微粒测试燃烧器”(以下称作“PTC”)，其为 550,000-Btu/hr(约 161 kW)的粉煤(“PC”)燃烧单元，其用来产生燃烧烟道气性能以及飞灰，用以代表大规模动力锅炉所产生的那些物质。使燃烧器垂直定向以使壁上沉积最小化。耐火衬里有助于保证完全燃烧的足够火焰温度，并阻止聚结或凝结的飞灰的迅速淬灭。基于表观气体速度，粒子在燃烧器中的平均滞留时间为大约 3 秒钟。PTC 的煤炭喷嘴从燃烧器的底部轴向地向上燃烧，以紊流混合将次级空气同心式地引入初级空气中。经由螺旋进料机和喷射器将煤炭引入到初级空气流中。使用电空气预热器精确控制燃烧空气温度。最初，PTC 使用冷水环形热交换器对集尘室(也指的是“织物过滤器”)或静电沉降器(ESP)提供烟道气的温度控制。然而，对从热交换器中收集到的灰分

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沉积物的分析表明，一些汞积聚在管道壁上。为了使这种影响最小化，对热交换器进行改进以提供更高的管道壁温度。

PTC 仪器设备容许连续地监测系统温度、压力、流速、烟道气成分浓度以及微粒控制装置(集尘室、高级混合式粒子收集器/AHPC™ 和/或静电沉降器/ESP)的操作数据并用数据记录器记录。

PTC 步骤

烟道气样本的选取为三个可取的系统样本点—炉子出口、粒子控制装置入口、粒子控制装置出口—中两个的组合。将烟道气通过样本调节装置以去除水分之后，通常对烟道气中的 O_2、CO、CO_2、SO_2 和 NO_x 进行分析。通常使用两个分析仪同时在炉子出口和粒子控制装置出口对每种成分进行分析。连续记录所有仪器上的浓度值。此外，以设定的时间间隔手动记录数据。使用一对 Rosemount Analytical NO_x 化学发光分析仪确定 NO_x。使用一对 Ametek Instruments 光度气体分析仪测定 SO_2。通过一对 Rosemount Analytical 多气体连续排放监测器对其余的气体进行测定。定期校准及维护各自的这些分析仪，从而提供准确的烟道气浓度测量。

集尘室容器为 20 英寸(50.8 cm)(ID)热跟踪及隔绝的腔室，引入的烟道气在底部附近。在 300°F(约 150℃)下燃烧器生成约 200 ACFM(实际立方英尺每分钟；约 5.7 实际 m^3/min)的烟道气，因此，三个 13 英尺 x5 英寸(3.96 m x12.7 cm)的袋子提供 4 英尺/min (1.22 m/min)的气布比。每个袋子随其自身的隔膜脉冲阀的操作而单独地进行清洁。为了对不同测试条件下压降的差值定量化，袋子的清洁是基于时间的，而不是基于压降引发的清洁周期。一旦引发袋子的清洁，以快速的顺序性在线对所有三个袋子进行脉冲。

还用单线式管状 ESP 代替织物过滤器进行了测试。设计 ESP 单元以使在 300°F(150℃)下提供的比收集面积为 125。因为 PTC 的烟道气

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流速为 130 SCFM(标准立方英尺每分钟；约 3.7 NCMM (标准 m^3/min))，则通过 ESP 的气体速度为 5 英尺/分钟(约 1.52 m/min)。ESP 单元的板距为 11 英寸(27.9 cm)。ESP 的设计要有利于各测试之间的彻底清洁，从而使所有的测试能具有相同的基础。

PTC 结果

结果例示于图 6 中。在图 6 中观察到，即使测试的吸附剂粒度显著地大于正规的吸附剂粒子，所处理的大于正规尺寸(即>60 微米)的活性碳吸附剂对于汞的捕集也相当有效。当在中试规模 ESP 前注入更大尺寸处理的 AC 时，大约 75%的汞被捕集，而当在中试规模织物过滤器(“FF”)前注入时，大约 85%的汞被捕集。注意在图 6 中(以及通篇中)，“Macf” (及“MACF”)表示百万实际立方英尺(1 MACF 为约 0.028 百万实际立方米或“MACM”)。

现在参考图 7，可以观察到，在相同的配置下，当在中试规模 ESP(菱形记号)前注入时，更大尺寸处理的 AC 表现出的性能优于更细的标准 AC(三角形)。作为比较，当在织物过滤器(FF) 前注入时，更大尺寸处理的 AC(方形)表现出从类似到稍微更差的性能。然而，对于这种应用而言，更大尺寸处理的 AC 可以从更小的飞灰粒子中物理地分离出来，然后可以将吸附剂再生、再循环及重新使用。这将充分地改善总利用性及经济性。这些数据因而说明，大于正规尺寸的吸附剂可以提供有效的汞控制，飞灰与 AC 的分离也容易，因此可以保留飞灰的销售及有益用途的特性。相应地，由于已经成功地证明了>60 μm 的吸附剂粒子，因而使用>40 μm 的粒子可以获得优异的汞控制，依照所使用的吸附剂粒子/灰分分离系统的情况，在一些应用中使用 40 μm 的粒子是优选的。注意在图 7 中(以及通篇中)，“Macf” (及“MACF”)表示百万实际立方英尺。

实施例 7-液相(有机溶剂)卤化

将 50 g 溴小心地加入到 1 升冷的轻石油中，制备溴在轻石油中的

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5%溶液。将 1 kg 的活性碳加入到大金属罐里的溴溶液中。在加入的期间用大桨叶片搅拌浆料，经短时间后，直到红色消失表明所有的溴已经与碳反应。用布氏漏斗真空过滤浆料。将过滤器中收集到的碳饼在110℃炉中干燥若干小时，直到其显示为干燥并达到恒重。如在实施例 1 中那样，碳中还仍然留有一定湿度。然后使干燥的碳在旋转桶中翻转，利用金属片破碎及疏解碳。

实施例 8-促进的活性碳吸附剂

采用基于上面描述的实验室规模的步骤来测试若干促进的活性碳吸附剂的初始活性及容量，所述吸附剂使用粉状碳，包括由商业上可得到的吸附剂制备的含溴活性碳和气凝胶碳膜吸附剂，以及初始前体碳用于比较。将粉状活性碳前体浸渍于搅拌的溶液中制备溴处理的碳，所述搅拌的溶液是溴在四氯化碳或二氯甲烷或者在 HBr 水溶液中的溶液，接着在环境温度下于空气中干燥，在 100℃炉中于空气或氮气氛下干燥。还可以使气体流过活性碳前体的旋转干燥床，从而从气相中浸渍溴来制备溴处理的碳。结果表明在溶液中加入次级组分改善了吸附剂的容量。

碳的初始测试在加热床上进行，其中包含元素态汞(11 μg/m^3)的合成烟道气流通过该床。流出气体中总的及元素态 Hg 的浓度用 Sir Galahad 汞 CEM(“连续排放监测器”)(制造商 P S Analytical，Deerfield Beach，FL，USA)来确定。在测试期间粉状吸附剂负载在石英过滤器上，以三层形式测试其它吸附剂。对初始的商业级粉状碳吸附剂、与该吸附剂在用 0.1 N HBr 处理之后离心收集粉末并干燥作了比较，表明汞捕集的活性从在入口处约 50% Hg 的初始捕集效率增大至 100%捕集。吸附剂在随后用 HBr 再生的比较表明，其不仅能够以与以前同样(100% 捕集)的水平捕集汞，而且其容量还延长若干分钟，因此是增强了的。如上面所描述的那样在溶液中或在干燥床中用分子态溴处理碳膜和碳纤维吸附剂得到类似的结果。

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实施例 9-流化/沸腾床制备

通过使气体流过与碳的流化床或沸腾床串联的液体溴储槽的方式，由包含分子态溴的气体组合物来进行分子态溴的浸渍，通过如此处理碳来制备活性碳吸附剂。依照所使用的比例，每 100 g 活性碳中碳吸收的溴量范围从(在一个实施例中的)<1 至约 30 g。

实施例 10-大规模测试

在该实施例中，使用集尘室(织物过滤器)或 ESP 来收集大规模商业粉煤燃烧设备的废气中的微粒。同时使用涤气器和吸附剂床从烟道气流中将不期望的成分在被送料到烟道之前清除掉。在该实施例中，在锅炉之后将卤素/卤化物促进的碳吸附剂注入到烟道气内。然而，一般来讲，可以将本发明的吸附剂注入到期望的位置(例如，在锅炉之前、之后或之内)。

在燃烧褐煤的设备上进行的一个示范测试中，烟道气相中汞(元素态) 浓度为 10 至 11 μ g/m^3。在 350℉至 375℉(约 175-190℃)下在集尘室中收集灰分和注入的碳。以 1.0 lb/MACF 的速率("MACF"和"Macf"代表百万实际立方英尺；1.0 lb/MACF 为约 16 kg/MACM(百万实际立方米))注入商业级活性碳粉末(未处理的)，得到 3.8-4.2 μ g/m^3 的汞流出浓度结果(分别代表气体中 62%-58%的汞清除)，速率 2.0 lb/MACF(约 32 kg/MACM)时，给出 74%-71%的清除。以 1.0 lb/MACF 速率注入溴处理的碳给出 73%-69%的清除结果，速率 2.0 lb/MACF 给出 86%-84%的清除。因此，使用本发明的溴促进的碳吸附剂表现出汞捕集的显著增加。

实施例 11A-任选的碱性组分的加入—实验室规模

为了捕集可能从吸附剂中释放的任何被氧化的汞，或者捕集烟道气中可能对吸附剂容量具有不利影响的某些硫或硒的氧化物，通过使用碱性物质与活性碳的共注入可以明显地改善活性碳的汞捕集效率。

制备由 37 mg 溴化的活性碳与 113 mg 氧化钙混合组成的过滤器，

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进行实验室规模测试。如在实施例 1 中描述的那样进行测试，并与带有惰性稀释剂的相同碳吸附剂进行比较。溴化的(2%) NORIT Darco FGD 吸附剂与惰性砂的混合物的穿透曲线示于图 8 中，与 CaO 的混合物的穿透曲线示于图 9 中。可以看到，与 CaO 的混合物的 50%穿透点增至 65 分钟，而与砂的混合物仅为 48 分钟。

实施例 11B-任选的碱性组分的加入—中试规模

在参考上面实施例 6 描述的中试规模的 PTC 燃烧器上进行测试，燃烧 Texas 褐煤，通过在织物过滤器上游共注入标准活性碳(本文也称为“AC”）和碱性物质来评价汞控制。典型结果例示在图 10 中。如图 10 所示，石灰与活性碳的共注入大大地增加了汞的清除。使用共注入吸附剂达到了大约 90%的汞清除，而即使在高得多的注入速率下，单独使用标准 AC 时只达到不到 60%的清除。类似测试的数据显示，注入类似量的碳酸钠与 AC 以及石灰与 AC，分别给出大约 80%以及 87%的汞清除结果。这些数据暗示，也可以用其它碱与 AC 共注入来改善汞的清除。其它数据显示，烟道气温度可以影响碱加入的有效性。进一步的测试数据表明，烟道气污染物、烟道气成分(SO_2、NO_x、HCl 等)、操作温度、汞的形式以及汞的浓度可以影响到碱加入的有效性。这表明理想的是能够原位地调节及定制碱与 AC 的比率，从而能在设定的原位条件下使清除最优化。

虽并不打算束缚于任何特定的理论，但可以如下地解释当两种物质共注入时在性能改善当中观察到的协同。首先，测试表明，AC 上的结合位置可以被硫物质和其它污染物所消耗。碱材料与这些物质相互作用及反应，从而使它们对于 AC 上汞结合位置的消耗最小化。其次，其它工作已经表明，即使结合位置完全被消耗，标准 AC 也将继续进行汞的氧化。然后该被氧化的汞可以与碱性物质反应，并随后被微粒控制装置捕集。因此，碱与处理的和/或未处理的 AC 的结合将协同地利用这两种机理，导致在降低的成本下的汞捕集的改善。

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实施例 12-用于气化燃料气体的溴化处理碳吸附剂 5% Br2W-AC 的制备

采用类似于实施例 3 中的步骤，制备溴在水中的 2.5 wt/vol % 溶液。将粒状的 Calgon F400 加入到溴溶液中，得到 5 wt/wt %溴化的碳产物。在加入期间及之后用大桨叶片搅拌溴溶液，直到水中的红色消失。用大布氏漏斗真空过滤悬浮液。滤饼在空气中干燥，然后在 110 ℃炉中干燥，直到获得稳定的重量，湿度减少到 15%。

5% Br2D-AC 的制备

如在实施例 7 中所描述的那样由在溶剂中加入 Br_2 制备溴化的吸附剂，不同的是，使用二氯甲烷代替轻石油作为溶剂，使用粒状的 Calgon F400。

5% PBr3-AC 的制备

采用实施例 5 中描述的方法由 PBr_3 制备磷卤化的吸附剂，不同的是，使用粒状的 Calgon F400。

氢气氛下的测试—步骤

为了模拟从煤炭气化的合成气或受热燃料气中捕集汞，采用使氮中包含 10% vol/vol 氢的气流以 500 cc/min 速率通过吸附剂来进行测试。气流中包含来自市售汞渗透源的 26.9 微克/m^3 的元素态汞。

在测试中，将吸附剂(0.5 g) 置于 0.39 英寸(1 cm, 内径)的玻璃管中，所述玻璃管配有中等熔块(medium frit)烧结玻璃滤片，从而在气流中保持吸附剂。包含吸附剂床的管与气体入口管连接，用于引入包含汞蒸汽的气流，在管连接的出口与检测器连接。检测器为 Semtech 2000 连续汞排放监测器。在环境温度下使管在氮气流(450 cc/min)中平衡 5 分钟以使系统稳定化。检测器显示在吸附剂床的流出物中汞的浓度为 0。(没有吸附剂的空白运转时的读数为 26.9 微克/m^3)。然后在选定的温度下将管置于炉中进行测试(250℃至 400℃)。收集检测器中的流出汞浓

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度数据，直到检测器显示恒定的读数有 5 分钟。然后在气流中加入氢(50 cc/min)，每 5 min 读取检测器的读数。根据温度及吸附剂的情况，在达 3 小时时段的不同时间段中，在几个炉温下进行测试。如在实施例 1 中那样，元素态汞浓度数据绘制为入口汞浓度的百分数对时间的图形。流出物中所有的汞为元素态的，因此单一检测器就足够了，无需用 $SnCl_2$ 捕集器来转化至元素态汞(如在实施例 1 中那样)。然后由穿透曲线确定 50%穿透的时间(达到 50%捕集的时间)。

结果

对于未溴化的吸附剂(Calgon F-400)、溴化的吸附剂(5% Br2W-AC 和 5% BrD-AC) 及磷溴化的吸附剂(5% PBr3-AC)，结果示于(以下)表 1 中。对于表 1 最后一栏中表示的时间段而言，每次运行在流出物中得到的最大汞浓度也记录在表 1 中。

在还原的氢条件下，未溴化的吸附剂立即穿透，仅 6.5 min 之后就耗尽。这种彻底失效的出现是因为氢在 100℃以上的任何温度下使未溴化的吸附剂中捕集的汞还原。两种溴化的吸附剂在高至至少 400℃的所有温度下均展示出极好的活性和良好的容量。磷溴化的吸附剂在高至至少 400℃的所有温度下展示出优异的活性和容量。

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表 1

50%穿透的时间

观察到的吸附剂最大 Hg 浓度

(10% 氢流)

吸附剂	温度(℃)	50%穿透(min)	最大[Hg] ($\mu g/m^3$)	时间(min)
F-400	250	6	20.3	6.5
5% Br2W-AC	250	>150	1.4	150
5% Br2W-AC	300	>180	4.3	180
5% Br2W-AC	350	160	15.1	180
5% Br2W-AC	400	60	13.9	65
5% PBr3-AC	250	>140	0.4	140
5% PBr3-AC	300	>150	0.5	150
5% PBr3-AC	350	>150	1.4	150
5% Br2D-AC	350	>180	2.1	180
5% Br2D-AC	400	>180	10.9	180

虽然已经给出和描述了本发明优选的实施方案，本领域的技术人员在不偏离本发明的实质和教导的情况下可以对其作出各种修改。本文中描述的实施方案仅为示范性的，不意味着具有限制性。文中公开的本发明的种种变化及修改也是可行的，也在本发明的范围之内。因此，保护范围不限于上面说明书给出的内容，而仅由随后的权利要求所限定，该范围包括权利要求主题的所有等同方案。

公开文本中提供的实施例仅为例示和解释的目的，不意味着对权利要求或本发明实施方案的限制。虽然已经给出和描述了本发明优选的实施方案，本领域的技术人员在不偏离本发明的实质和教导的情况下可以对其作出各种修改。基于本文的公开内容，本领域的技术人员可以很容易地确定出本发明给定实施中的方法标准、设备等。本文描述的实施方案仅为示范性的，不意味着具有限制性。文中公开的本发明的多种变化及修改也是可行的，也在本发明的范围之内。关于本发

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明中的任何要素使用的术语“任选的”意味着表明该主题要素是需要的或者不需要的。两种选择方案均意味着落于本发明的范围之内。

背景部分中对参考文献的讨论并不是承认其为本发明的在先技术，特别是公布日期可能在本申请的优先权日期之后的任何参考文献。据此将本文引用的所有专利、专利申请及出版物的公开内容全部全文引入到本文中以供参考，其引入的范围对本文中的陈述内容提供示范性的、程序性的或其它详细的补充。

虽然本文描述的本发明为吸附剂材料以及其制备和用途的相关方法，但并不意味着局限于所描述的细节，在不偏离本发明实质的情况下可以做出各种修改及结构变化，这都在权利要求的等同方案的范围之内。

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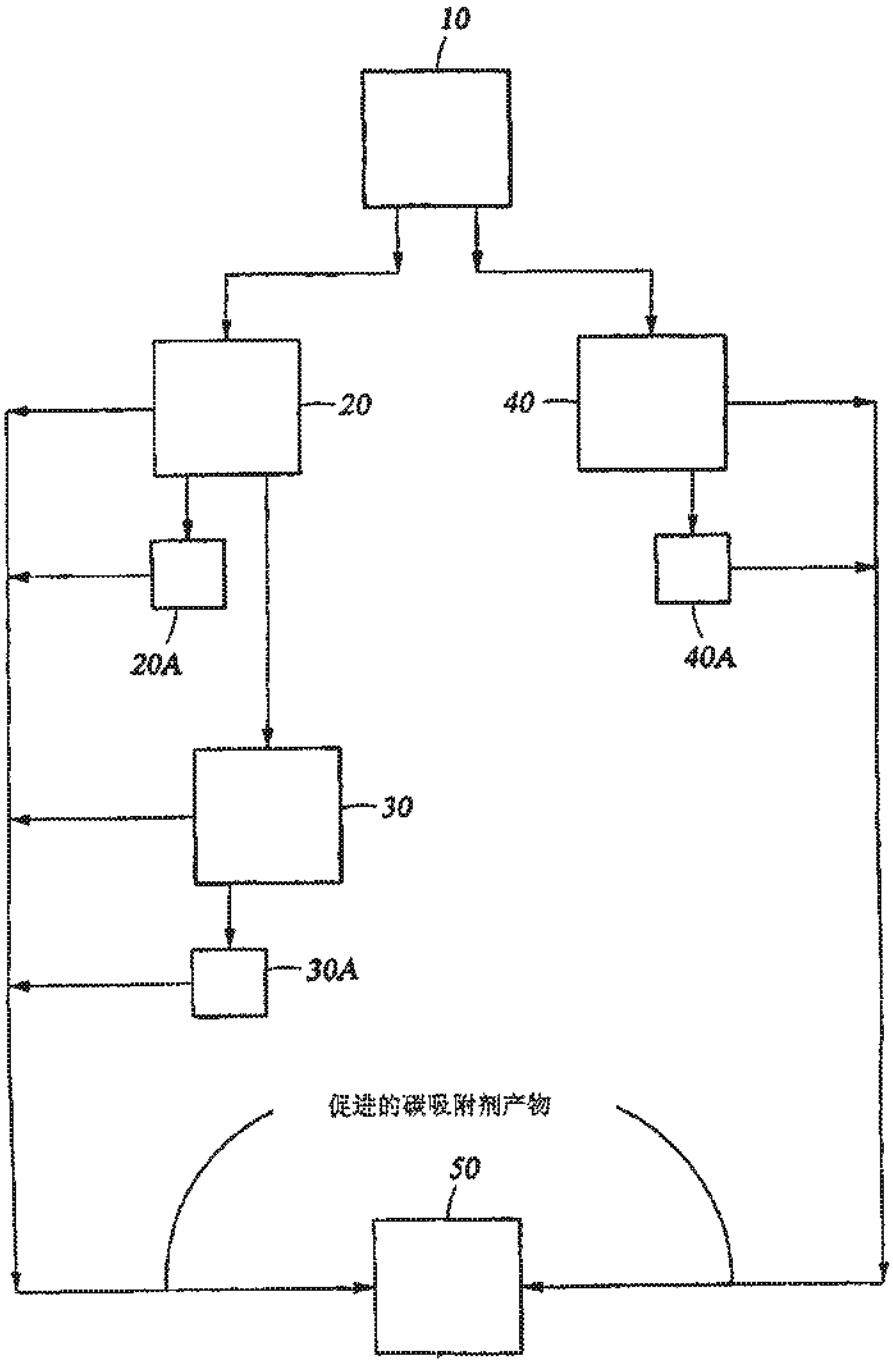


图1

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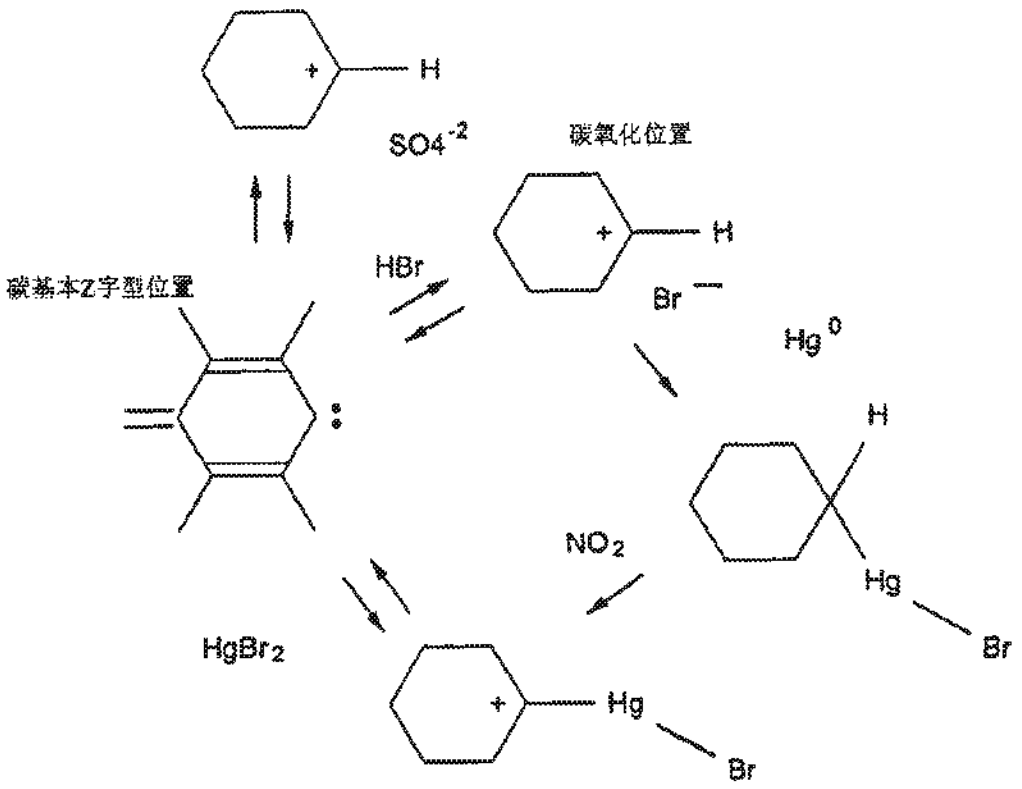


图2

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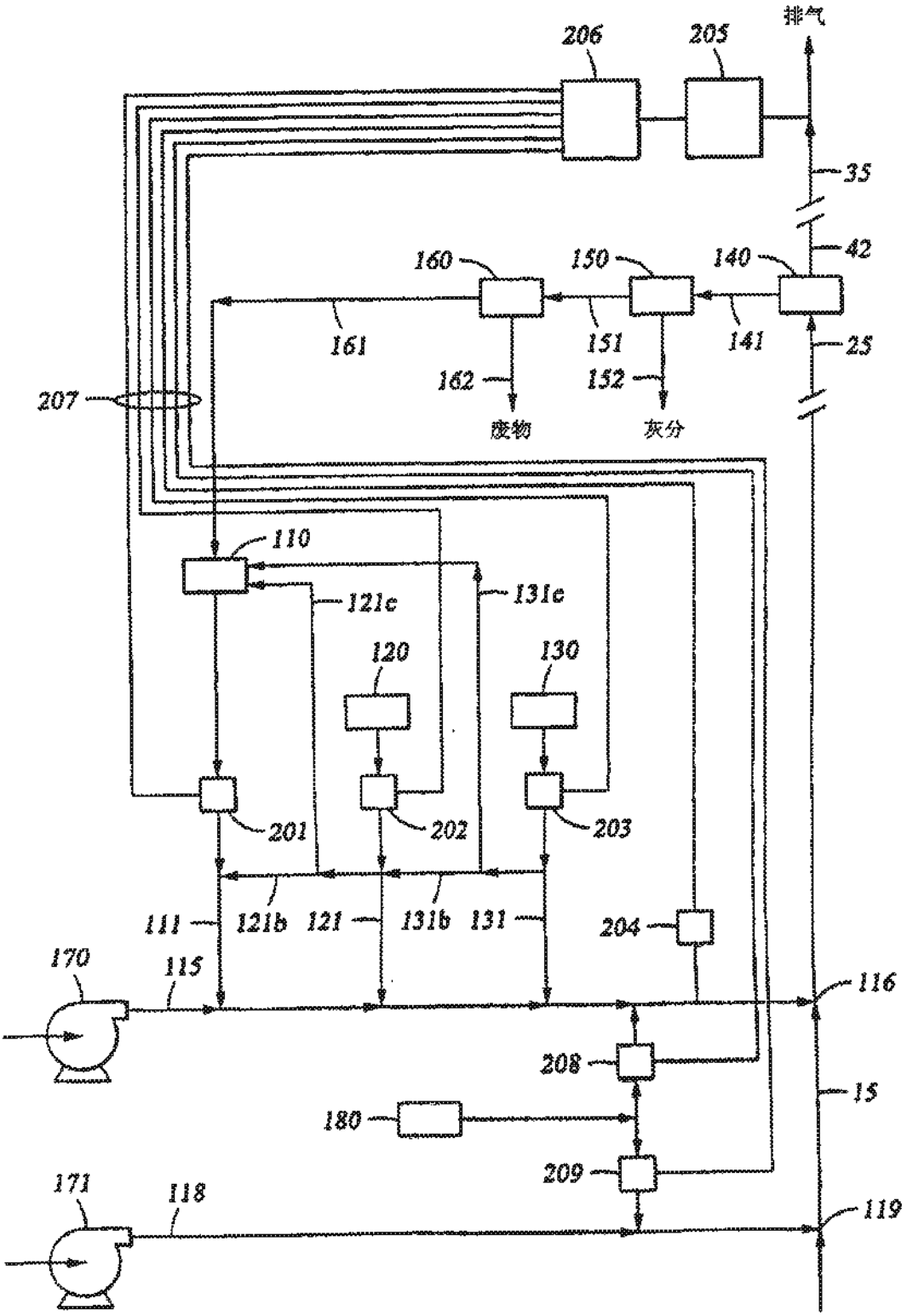


图3

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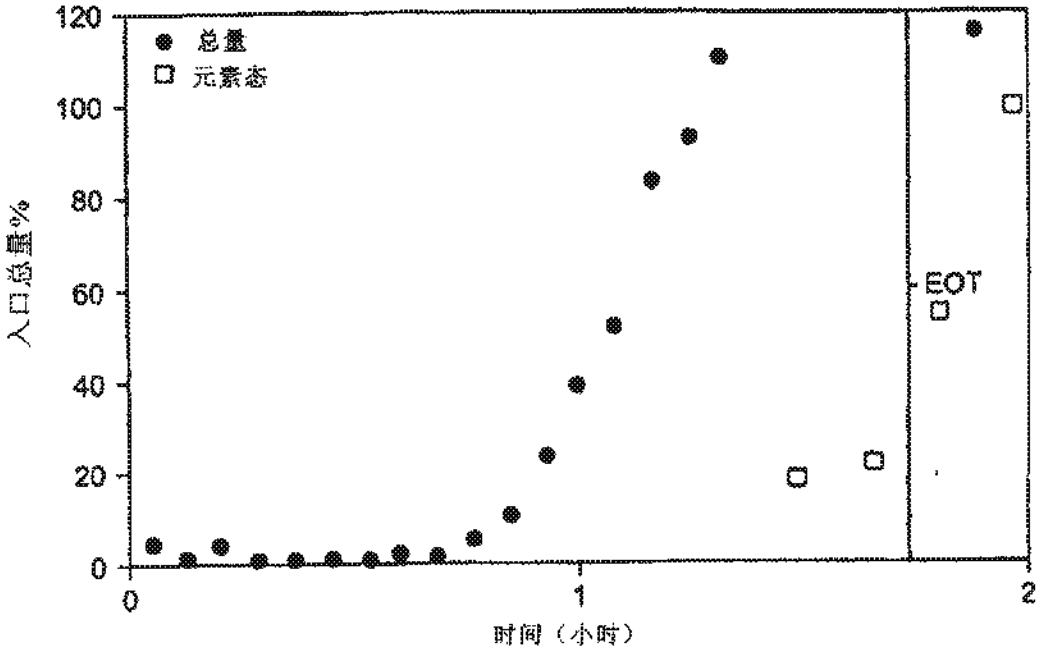


图4

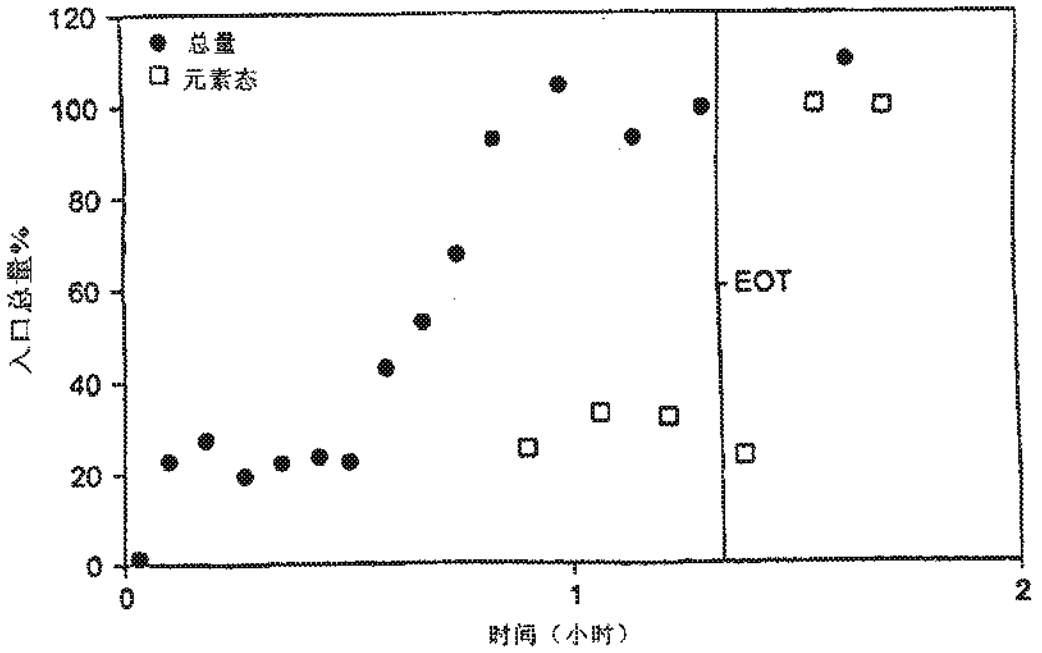


图5

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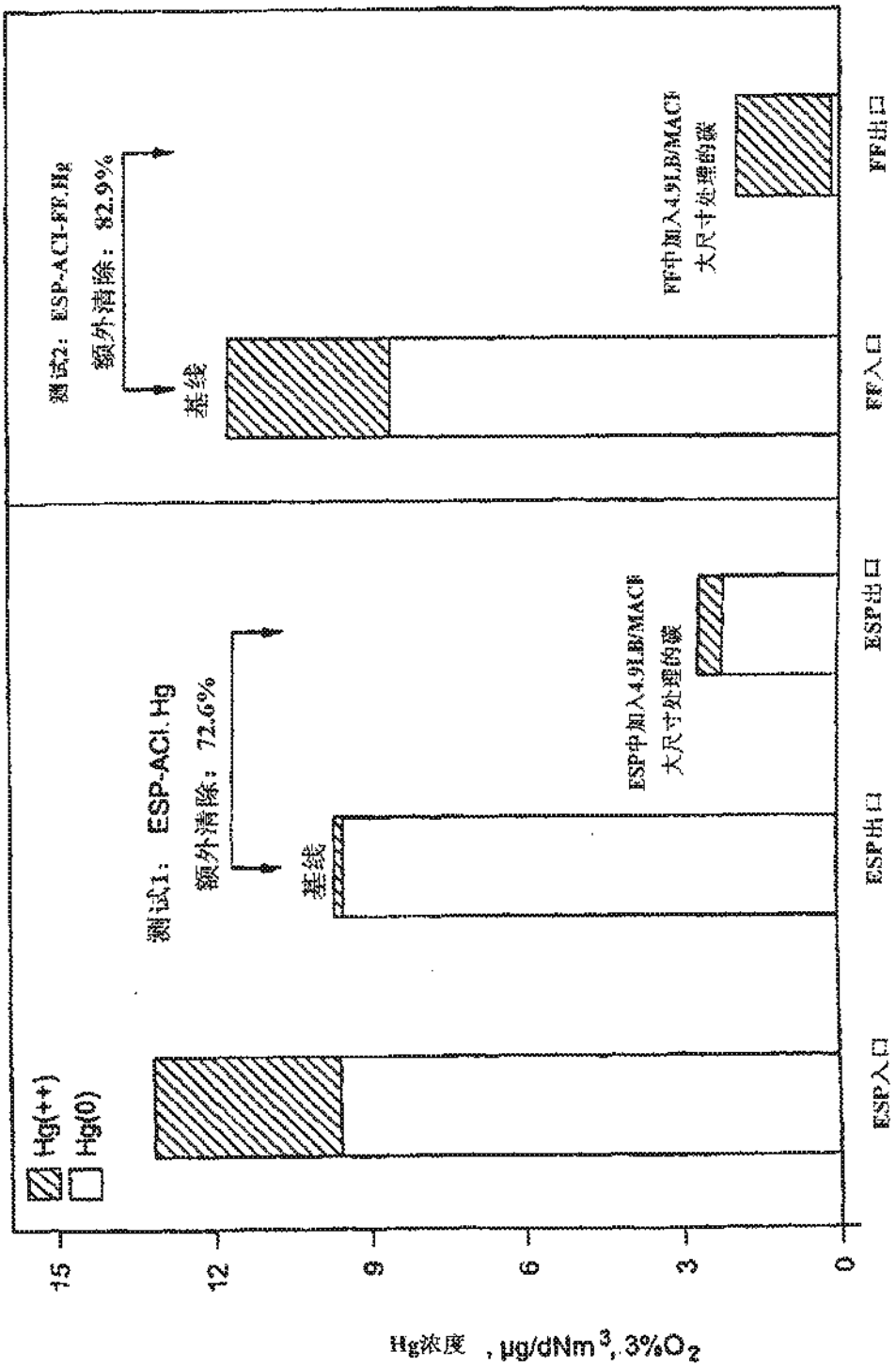


图6

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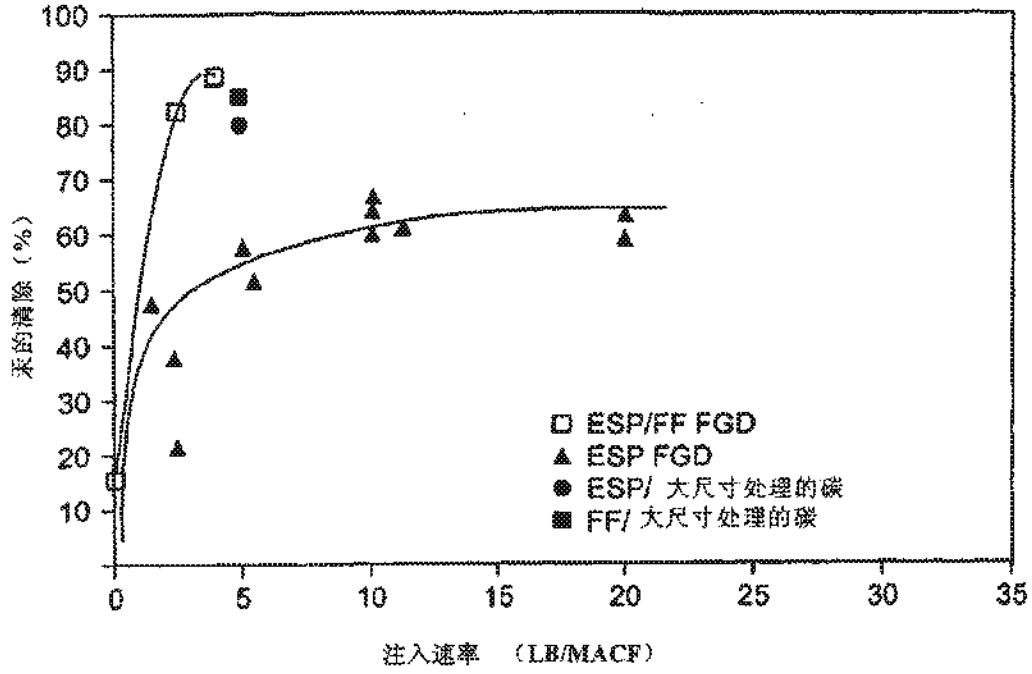


图7

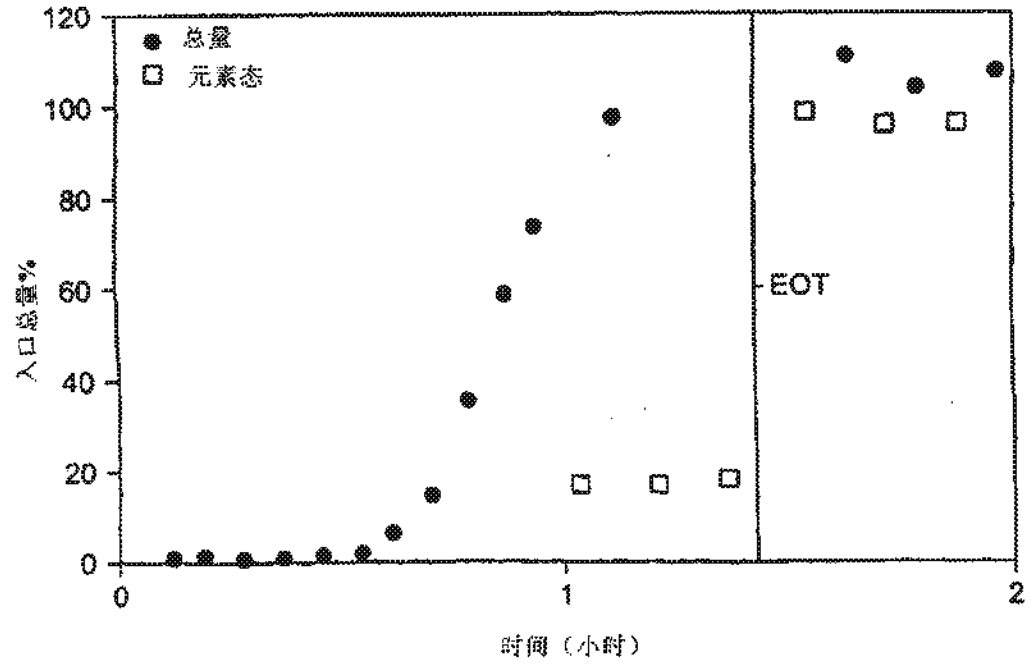


图8

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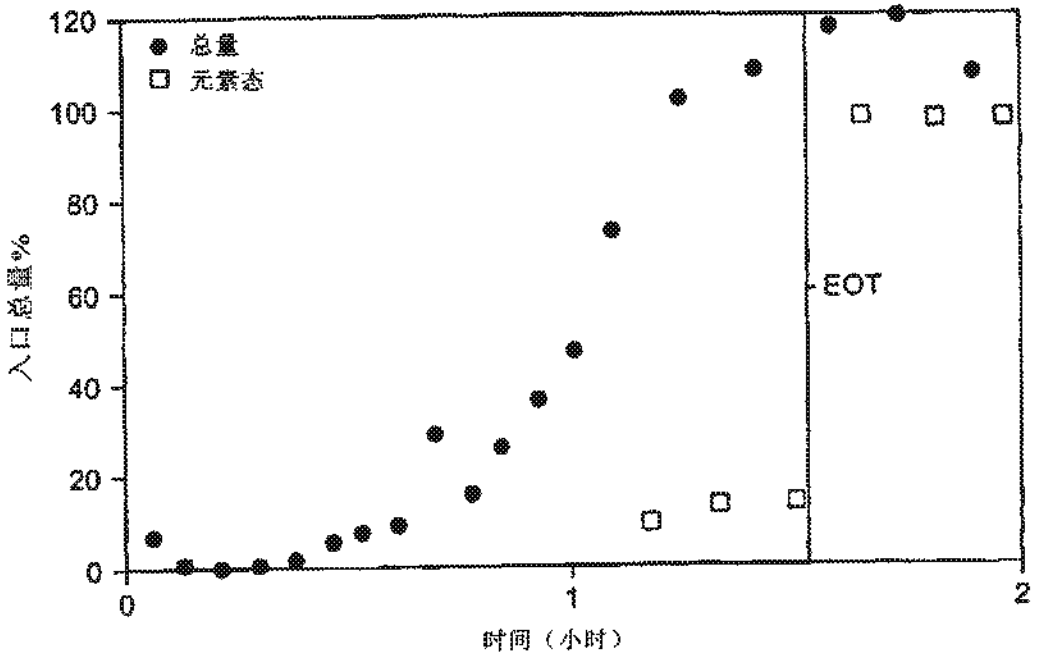


图9

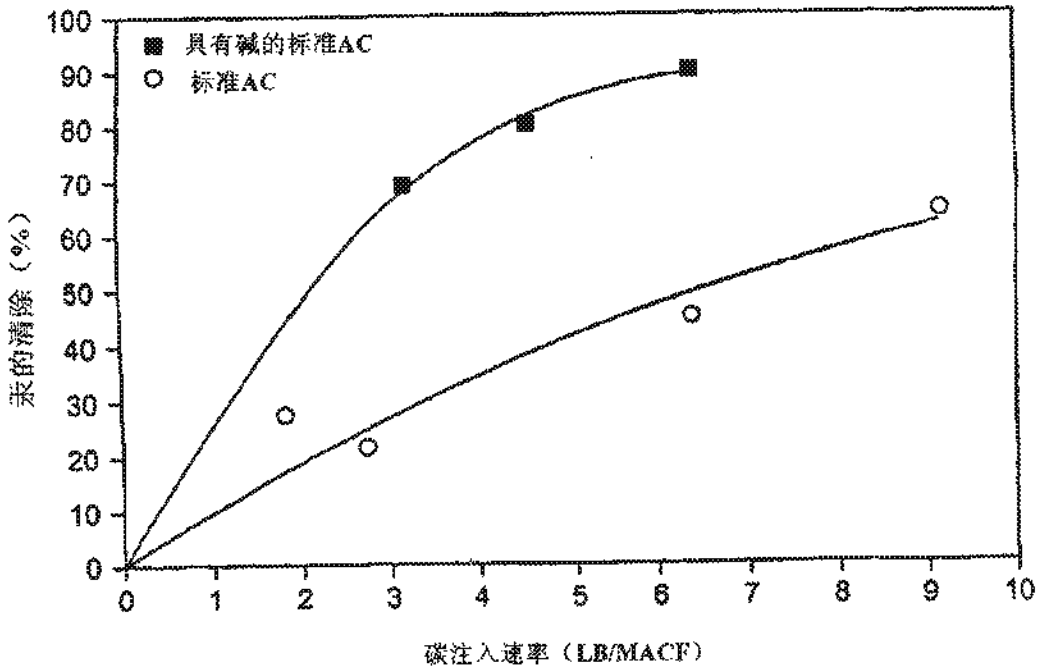


图10

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Espacenet

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Organic complex supported activated carbon adsorbent and its preparation method and application

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Abstract of CN101053820 (A)

The invention relates to an organic complex supported activated carbon adsorbent, which has activated carbon as a carrier and organic complex distributed on a pore path of the activated carbon. The preparing method comprises: immerging the activated carbon treated by vacuumizing in a maceration extract; removing solvent by vacuumizing; leaching the sample using an eluate while vacuum filtrating; and washing the the sample using deionized water while vacuum filtrating. The adsorbent is applicable for purifying peroxide (H2O2) solution by a way of: adding the adsorbent into the peroxide solution, absorbing and filtering the mixture to effectively remove organic matter impurity from the peroxide solution as well as remove various metallic iron impurities such as Al, Ti, Cr, Mn, Fe, Ni, Cu, Zn, Ga, Rb, Sr, Mo, Ag, Sb, W, Au, Hg, etc., thus the aim of purification is achieved.; Accordingly, the adsorbent of the invention is suitable for purifying of peroxide solution, in particular commercial grade peroxide solution having more organic matter impurity.

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权利要求书 2 页 说明书 9 页

[54] 发明名称

有机络合物负载型活性炭吸附剂及其制备方法和应用

[57] 摘要

本发明涉及一种有机络合物负载型活性炭吸附剂，是以活性炭为载体，有机络合物主要分布在活性炭的孔道上；其制备方法包括：经抽真空处理的活性炭浸没在浸渍液中；抽真空脱除溶剂；用淋洗液对样品边淋洗边进行真空抽滤；用去离子水对样品边冲洗边进行真空抽滤；所述吸附剂可用于纯化过氧化氢(H_2O_2)溶液，即将吸附剂加入到过氧化氢溶液中，进行吸附，过滤，可有效地脱除过氧化氢溶液中的有机物杂质，以及可对 Al、Ti、Cr、Mn、Fe、Ni、Cu、Zn、Ga、Rb、Sr、Mo、Ag、Sb、W、Au、Hg 等多种金属离子杂质进行同时脱除，从而达到净化的目的；本发明的吸附剂适用于过氧化氢溶液，特别是含有机物杂质较多的工业级过氧化氢溶液的纯化。