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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
12/201,595	08/29/2008	Edwin S. Olson	16291.3USD1	9735

23552 7590 10/01/2010

MERCHANT & GOULD PC
P.O. BOX 2903
MINNEAPOLIS, MN 55402-0903

EXAMINER	
ORLANDO, AMBER ROSE	
ART UNIT	PAPER NUMBER
1797	

MAIL DATE	DELIVERY MODE
10/01/2010	PAPER

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

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

Notice of Abandonment	Application No.	Applicant(s)	
	12/201,595	OLSON ET AL.	
	Examiner	Art Unit	
	AMBER ORLANDO	1797	

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address--

This application is abandoned in view of:

1. ☒ Applicant's failure to timely file a proper reply to the Office letter mailed on *10 March 2010*.
   (a) ☐ A reply was received on _____ (with a Certificate of Mailing or Transmission dated _____), which is after the expiration of the period for reply (including a total extension of time of _____ month(s)) which expired on _____.
   (b) ☐ A proposed reply was received on _____, but it does not constitute a proper reply under 37 CFR 1.113 (a) to the final rejection.
   (A proper reply under 37 CFR 1.113 to a final rejection consists only of: (1) a timely filed amendment which places the application in condition for allowance; (2) a timely filed Notice of Appeal (with appeal fee); or (3) a timely filed Request for Continued Examination (RCE) in compliance with 37 CFR 1.114).
   (c) ☐ A reply was received on _____ but it does not constitute a proper reply, or a bona fide attempt at a proper reply, to the non-final rejection. See 37 CFR 1.85(a) and 1.111. (See explanation in box 7 below).
   (d) ☒ No reply has been received.

2. ☐ Applicant's failure to timely pay the required issue fee and publication fee, if applicable, within the statutory period of three months from the mailing date of the Notice of Allowance (PTOL-85).
   (a) ☐ The issue fee and publication fee, if applicable, was received on _____ (with a Certificate of Mailing or Transmission dated _____), which is after the expiration of the statutory period for payment of the issue fee (and publication fee) set in the Notice of Allowance (PTOL-85).
   (b) ☐ The submitted fee of $_____ is insufficient. A balance of $_____ is due.
   The issue fee required by 37 CFR 1.18 is $_____. The publication fee, if required by 37 CFR 1.18(d), is $_____.
   (c) ☐ The issue fee and publication fee, if applicable, has not been received.

3. ☐ Applicant's failure to timely file corrected drawings as required by, and within the three-month period set in, the Notice of Allowability (PTO-37).
   (a) ☐ Proposed corrected drawings were received on _____ (with a Certificate of Mailing or Transmission dated _____), which is after the expiration of the period for reply.
   (b) ☐ No corrected drawings have been received.

4. ☐ The letter of express abandonment which is signed by the attorney or agent of record, the assignee of the entire interest, or all of the applicants.

5. ☐ The letter of express abandonment which is signed by an attorney or agent (acting in a representative capacity under 37 CFR 1.34(a)) upon the filing of a continuing application.

6. ☐ The decision by the Board of Patent Appeals and Interference rendered on _____ and because the period for seeking court review of the decision has expired and there are no allowed claims.

7. ☒ The reason(s) below:

A call was placed to the applicant's attorney of record Dennis Daley on September 29, 2010 to confirm the abandonment status of the application.

/Walter D. Griffin/
Supervisory Patent Examiner, Art Unit 1797

Petitions to revive under 37 CFR 1.137(a) or (b), or requests to withdraw the holding of abandonment under 37 CFR 1.181, should be promptly filed to minimize any negative effects on patent term.

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Office Action Summary	Application No.	Applicant(s)	
	12/201,595	OLSON ET AL.	
	Examiner	Art Unit	
	AMBER ORLANDO	1797	

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE *3* MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

1)☒ Responsive to communication(s) filed on *01 December 2009*.

2a)☒ This action is **FINAL**. 2b)☐ This action is non-final.

3)☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims

4)☒ Claim(s) *1-3,7,10,14-16,34 and 35* is/are pending in the application.

4a) Of the above claim(s) _____ is/are withdrawn from consideration.

5)☐ Claim(s) _____ is/are allowed.

6)☒ Claim(s) *1-3,7,10,14-16,34 and 35* is/are rejected.

7)☐ Claim(s) _____ is/are objected to.

8)☐ Claim(s) _____ are subject to restriction and/or election requirement.

Application Papers

9)☐ The specification is objected to by the Examiner.

10)☐ The drawing(s) filed on _____ is/are: a)☐ accepted or b)☐ objected to by the Examiner.

Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).

Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).

11)☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

Priority under 35 U.S.C. § 119

12)☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

a)☐ All b)☐ Some * c)☐ None of:

1.☐ Certified copies of the priority documents have been received.

2.☐ Certified copies of the priority documents have been received in Application No. _____.

3.☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

* See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

1) ☐ Notice of References Cited (PTO-892)
2) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
3) ☒ Information Disclosure Statement(s) (PTO/SB/08) Paper No(s)/Mail Date _____.
4) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date. _____ .
5) ☐ Notice of Informal Patent Application
6) ☐ Other: _____.

DETAILED ACTION

This action is in response to the correspondence filed 12/01/2009.

Claims 1, 3, 7, 10, 14, 15, and 16 have been amended.

Claims 4-6, 8, 9, 11-13 and 1-33 have been cancelled.

Claims 34 and 35 are new.

Claims 34 and 35 are objected to.

Claims 1-3, 7, 10, 14-16, 34 and 35 are rejected.

Claims 1-3, 7, 10, 14-16, 34 and 35 have been examined and are pending.

Claim Rejections - 35 USC § 102

The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

> A person shall be entitled to a patent unless –
>
> (a) the invention was known or used by others in this country, or patented or described in a printed publication in this or a foreign country, before the invention thereof by the applicant for a patent.

Claims 1, 34 and 35 are rejected under 35 U.S.C. 102(a) as being anticipated by Srinivasachar et al. US 6,848,374.

For claim 1, the Srinivasachar et al. reference discloses a promoted carbon sorbent comprising a base activated carbon that has been treated with a promoter selected from the group consisting of halides, halogens, and combinations thereof stream (column 6, lines 2-20).

The reference does not explicitly state the carbon sorbent and the promoter reacting forming a reaction product of the promoter and a carbene site on the base activated carbon to form a carbocation accompanied by a halide anion.

One having ordinary skill in the art at the time the invention was made would realize that contacting the activated carbon with the promoter would inherently produce the same results as that in the present invention (e.g. cause a reaction between the two, forming a reaction product of the promoter and a carbene site on the base activated carbon to form a carbocation). The sorbent being effective for the removal of mercury from a gas stream is the method of using the promoted carbon. The product disclosed by the Srinivasachar et al. reference would be able to be effective for the removal of mercury from a gas stream. Therefore the method of using the product does not further limit the claim.

For claims 34 and 35, the promoted carbon sorbent of claim 1 is disclosed above.

The sorbent oxidizes and captures mercury from a gas stream and being effective for the removal of at least one of boron, tin arsenic, gallium, antimony, lead, bismuth, cadmium, silver, copper and zinc is the method of using the promoted carbon. The product disclosed by the Srinivasachar et al. reference would be able to oxidize and capture mercury from a gas stream and being effective for the removal of at least one of boron, tin arsenic, gallium, antimony, lead, bismuth, cadmium, silver, copper and zinc is the method of using the promoted carbon. Therefore the method of using the product does not further limit the claim.

Claim Rejections - 35 USC § 103

The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

> (a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negatived by the manner in which the invention was made.

The factual inquiries set forth in *Graham* **v.** *John Deere Co.*, 383 U.S. 1, 148 USPQ 459 (1966), that are applied for establishing a background for determining obviousness under 35 U.S.C. 103(a) are summarized as follows:

1. Determining the scope and contents of the prior art.
2. Ascertaining the differences between the prior art and the claims at issue.
3. Resolving the level of ordinary skill in the pertinent art.
4. Considering objective evidence present in the application indicating obviousness or nonobviousness.

This application currently names joint inventors. In considering patentability of the claims under 35 U.S.C. 103(a), the examiner presumes that the subject matter of the various claims was commonly owned at the time any inventions covered therein were made absent any evidence to the contrary. Applicant is advised of the obligation under 37 CFR 1.56 to point out the inventor and invention dates of each claim that was not commonly owned at the time a later invention was made in order for the examiner to consider the applicability of 35 U.S.C. 103(c) and potential 35 U.S.C. 102(e), (f) or (g) prior art under 35 U.S.C. 103(a).

Claims 2, 10 and 15 are rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374 as applied in claim 1 above, and further in view of Reiss 5,575,982.

For claim 2, the Srinivasachar et al. reference discloses 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 (column 1, lines 37-45 and column 5, lines 20-29) and the promoter is selected from the group consisting of molecular halogens, Group V halides, Group VI halides, hydrohalides, and combinations thereof (column 6, lines 2-20). The reference does not disclose an average particle size greater than 40 micrometers

The Reiss reference discloses the activated carbon has an average particle size greater than 40 μm (Reiss, column 4, lines 59-65).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar reference to include the activated carbon has an average particle size greater than 40 μm (Reiss column 4, lines 59-65) because these particle diameters of the activated carbon can be effectively introduced into the exhaust gas stream and cause optimum adsorption.

For claims 10 and 15, the Srinivasachar et al. discloses a component comprising a halogen or a hydrohalide, a Group V halides, Group VI halides, and the component is substantially in gas form when combined with the base activated carbon (column 6, lines 2-20). The reference does not explicitly state the component being the secondary component. It would have been obvious to one having ordinary skill in the art at the time

the invention was made to have included an additional (e.g. secondary) component comprising a halogen because the reference already established that the mercury capacity is enhanced by adding a halogen (column 6, lines 32-39) and therefore one could assume that by adding another component of the same composition would further promote the removal of mercury from the gas stream. The claim does not state how the secondary component is included within the promoted carbon sorbent (e.g. is it reacting/bonding/coating). If it is not reacting/bonding/coating, then it is not part of the promoted carbon sorbent and does not further limit the claim.

Claim 3 is rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374 and Reiss 5,575,982 as applied in claim 2 above, and further in view of Moller et al. US 4,889,698.

For claims 3, the Srinivasachar et al. reference does not disclose the sorbent comprises an activated carbon matrix in a fixed or a moving bed structure and the structure consisting of moving fiber screens and monoliths.

The Moller et al. reference discloses using the sorbent comprises an activated carbon matrix in a fixed or a moving bed structure (column 4, lines 39-45). The reference does not explicitly state the structure is selected from the group consisting of moving fiber screens and monoliths. Merely choosing different types of fixed bed structures would be obvious to one having ordinary skill in the art.

It would have been obvious to one having ordinary skill in the art at the time the sorbent comprises an activated carbon matrix in a fixed or a moving bed structure and the structure consisting of moving fiber screens and monoliths (Moller et al. column 4,

lines 39-45) because this allows the contaminated gas to reach as much of the activated carbon as possible.

Claim 7 is rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374 and Reiss 5,575,982 as applied in claim 2 above, and further in view of Broderick US 6,942,840.

For claim 7, the Srinivasachar et al. reference does not disclose 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.

The Broderick reference discloses disclose 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 (column 4, lines 12-25).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar et al. reference to include disclose 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 (Broderick column 4, lines 12-25) because this allows the mercury to vented to the atmosphere without additional treatment.

Claim 14 is rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374.

For claim 14, the Srinivasachar et al. reference discloses the promoter is sprayed on the base activated carbon (column 6, lines 2-8). The reference does not explicitly state the promoter is substantially in gas phase. It would be well within the knowledge of

one having ordinary skill in the art to apply the promoter by spraying in any fluid form (including the gases form) therefore allowing the promoter the maximum amount of surface area of the base activated carbon.

Claim 16 is rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374 and Reiss 5,575,982 as applied in claim 10 above, and further in view of Moller et al. US 4,889,698.

For claim 16, the Srinivasachar reference discloses combining the promoter and the activated carbon (column 6, lines 2-20). The reference does not disclose injecting the optional secondary component and the promoter simultaneously. The promoters are essentially the same so therefore it would be obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar reference to include injecting the optional secondary component and the promoter simultaneously because this will cut down on production costs.

The Moller et al. reference discloses reacting the 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 (column 7, lines 43-56).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar et al. reference to include reacting the 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 (Moller et al. column 7, lines 43-56) because this promotes the removal of mercury from the gas.

Response to Arguments

1. Applicant's arguments filed 10/16/2009 have been fully considered but they are not persuasive.

2. For claim 1, the applicant argues that “Srinivasachar is directed at the production of a carbonaceous sorbent for the capture of elemental mercury”. In addition, *Srinivasachar* is directed at the "treatment of the carbonaceous sorbent to enhance the capture of elemental mercury." See *Srinivasachar* at column 2, line 10- 18. The treatment of the carbonaceous sorbent involves "condensing/adsorbing" a halogen 58 onto a sorbent 28. See *Srinivasachar* at column 6, lines 13-22, and figure 1. Because *Srinivasachar* describes "condensing/adsorbing" the halogen onto the sorbent, there is clearly no intention by *Srinivasachar* of reacting the halogen and the sorbent. Furthermore, *Srinivasachar* fails to describe conditions that provide for reacting a promoter and a carbene site on a base activated carbon to form a carbocation accompanied by a halide anion according to the presently claimed invention.”

3. For claim 1, the applicant is merely contacting two substances, the promoter and the sorbent. The Srinivasachar reference discloses contacting these same two substances, and therefore would result in the same product as the applicant’s current invention. The applicant must show what conditions are different within the applicant’s invention that would result in the combination of the same components providing differing results. The applicant is claiming that some how the outcome of these two substances contacting would be different without providing sufficient evidence on how this would occur.

4. The applicant also argues that "The conditions described by Srinivasachar simply provide for depositing a halogen on the surface of the sorbent particles contacted by an oxidizing agent".

5. The Examiner disagrees. The burden is placed on the applicant to show how the conditions (e.g. that would make the applicants current combination of sorbent and promoter interact) are different from that of the Srinivasachar, which result in the sorbent and the promoter interacting in a different way than that of the Srinivasachar reference.

6. The applicant contends that the Srinivasachar reference has not achieved a reaction between the halogen and the carbene site on the activated carbon, because the Srinivasachar reference would have seen a greater improvement. Furthermore the applicant argues that the Srinivasachar reference shows preference to using carbon black over activated carbon, and therefore Srinivasachar does not recognize an advantage of reacting halogens with a carbene site on base activated carbon.

7. The applicant appears to be claiming unexpected results when comparing the Srinivasachar combination of the halogen and the activated carbon to that of the applicant's invention. The applicant must show specific results, as well as how the combination of the same components results in differing results. Furthermore Srinivasachar reference discloses both the use of carbon black and activated carbon. The use of one over the does not provide evidence that the Srinivasachar reference does not show the promoter reacting forming a reaction product of the promoter and a carbene site on the base activated carbon to form a carbocation accompanied by a

halide anion. The applicant must provide evidence that there would be an improvement when using activated carbon compared with carbon black, when a reaction between the halogen and the carbene sites on the activated carbon.

Conclusion

8. Applicant's amendment necessitated the new ground(s) of rejection presented in this Office action. Accordingly, **THIS ACTION IS MADE FINAL**. See MPEP § 706.07(a). Applicant is reminded of the extension of time policy as set forth in 37 CFR 1.136(a).

A shortened statutory period for reply to this final action is set to expire THREE MONTHS from the mailing date of this action. In the event a first reply is filed within TWO MONTHS of the mailing date of this final action and the advisory action is not mailed until after the end of the THREE-MONTH shortened statutory period, then the shortened statutory period will expire on the date the advisory action is mailed, and any extension fee pursuant to 37 CFR 1.136(a) will be calculated from the mailing date of the advisory action. In no event, however, will the statutory period for reply expire later than SIX MONTHS from the date of this final action.

Any inquiry concerning this communication or earlier communications from the examiner should be directed to AMBER ORLANDO whose telephone number is (571)270-3149. The examiner can normally be reached on Mon.-Thurs. (6:30-5:00).

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Walter Griffin can be reached on (571) 272-1447. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

AO

/Walter D. Griffin/
Supervisory Patent Examiner, Art Unit 1797

EAST Search History

EAST Search History (Prior Art)

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
L1	65	423/210.ccls. and remov $4 with mercury and carbon and (halide or halogen)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2010/02/26 15:14
L2	71	95/134.ccls. and remov $4 with mercury and (V or Nb or Ta or Db or Cr or Mo or W or Sg or Group V or Group VI)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2010/02/26 15:14
L3	19	(remov$5 near3 mercury) and (carbon same in-flight)	US-PGPUB; USPAT	OR	OFF	2010/02/26 15:14
L4	1512	(remov$5 near3 mercury) and (carbon same in flight)	US-PGPUB; USPAT	OR	OFF	2010/02/26 15:14
L5	5	(remov$5 near3 mercury) and (carbon same in flight)	US-PGPUB; USPAT	ADJ	OFF	2010/02/26 15:14
L6	94	sorbent and mercury and (surface reaction)	US-PGPUB; USPAT; USOCR	ADJ	ON	2010/02/26 15:15
L7	37	110/345.ccls. and sorbent same mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2010/02/26 15:16
S1	4	2004/0003716	US-PGPUB; USPAT	OR	OFF	2007/07/26 10:53
S2	1	"20040003716"	US-PGPUB; USPAT	OR	OFF	2007/07/26 10:54
S3	8	(remov$5 near3 mercury) and (carbon same in-flight)	US-PGPUB; USPAT	OR	OFF	2007/07/24 10:18
S4	8	(remov$5 near3 mercury) and (carbon same in-flight)	US-PGPUB; USPAT	ADJ	ON	2007/07/24 10:48
S5	2	"6953494"	US-PGPUB; USPAT	ADJ	ON	2007/07/24 10:48

S6	42	("1788466" | "1984164" | "20010002387" | "20030161771" | "20040013589" | "2511288" | "3194629" | "3662523" | "3849267" | "3956458" | "3961020" | "4040802" | "4075282" | "4174373" | "4196173" | "4394354" | "4427630" | "4500327" | "4578256" | "4708853" | "4889698" | "4917862" | "5179058" | "5202301" | "5288306" | "5320817" | "5336835" | "5352647" | "5372619" | "5403548" | "5435980" | "5505766" | "5827352" | "6475461" | "6514907" | "6521021" | "6533842").PN. OR ("6953494").URPN.	US-PGPUB; USPAT; USOCR	OR	OFF	2007/07/24 10:48
S7	41	("1788466" | "1984164" | "20010002387" | "20030161771" | "20040013589" | "2511288" | "3194629" | "3662523" | "3849267" | "3956458" | "3961020" | "4040802" | "4075282" | "4174373" | "4196173" | "4394354" | "4427630" | "4500327" | "4578256" | "4708853" | "4889698" | "4917862" | "5179058" | "5202301" | "5288306" | "5320817" | "5336835" | "5352647" | "5372619" | "5403548" | "5435980" | "5505766" | "5827352" | "6475461" | "6514907" | "6521021" |	US-PGPUB; USPAT; USOCR	OR	OFF	2007/07/24 10:48

		"6533842").PN. OR ("6953494").URPN. and inflight				
S8	41	("1788466" | "1984164" | "20010002387" | "20030161771" | "20040013589" | "2511288" | "3194629" | "3662523" | "3849267" | "3956458" | "3961020" | "4040802" | "4075282" | "4174373" | "4196173" | "4394354" | "4427630" | "4500327" | "4578256" | "4708853" | "4889698" | "4917862" | "5179058" | "5202301" | "5288306" | "5320817" | "5336835" | "5352647" | "5372619" | "5403548" | "5435980" | "5505766" | "5827352" | "6475461" | "6514907" | "6521021" | "6533842").PN. OR ("6953494").URPN. and in-flight	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/24 10:53
S9	50	"4500327"	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/24 10:54
S10	9	in-flight near4 reaction	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 09:24
S11	13	in-flight same (mercury and remov$5)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/24 16:41
S12	0	carbon and (in flight) near4 reaction	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/24 16:42
S13	6	carbon and (in-flight) near4 reaction	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 09:21
S14	9	in-flight near4 reaction and in-flight	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 09:32

S15	5261	in-flight	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 09:33
S16	118	in-flight and mercury	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 09:33
S17	12	in-flight and (remov$4 with mercury)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 09:34
S18	0	In-Flight Modification Of Inorganic Particles Within A Reaction Product Flow	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 09:34
S19	7	In-Flight Modification	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 09:53
S20	4	In-Flight Capture	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 09:42
S21	19	(sorbent near3 composition) same (remov$4 near3 mercury)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 09:49
S22	252	(sorbent near3 Bromine OR iodine) same (remov $4 near3 mercury)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 09:49
S23	6	(sorbent near3 (Bromine OR iodine)) same (remov$4 near3 mercury)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 09:49
S24	0	In-Flight Modification and mercury	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 10:43
S25	9	(mercury near3 remov $4) same (surface reactions)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 10:44
S26	1227	sorbent and mercury and (surface reaction)	US-PGPUB; USPAT; USOCR	OR	OFF	2007/07/25 13:56
S27	43	sorbent and mercury and (surface reaction)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2007/07/25 13:56
S28	64	sorbent and mercury and (surface reaction)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 13:57
S29	7	sorbent and mercury and (surface reaction) and bromine	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 13:57

S30	10	sorbent and (remov$4 near3 mercury) and (surface reaction)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 14:55
S31	2	"5409522" and (surface reaction)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 14:02
S32	0	"5409522" and (surface reaction) and (iodine or bromine)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 14:03
S33	1	"20060048646" and in-flight	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 14:20
S34	165	sorbent and (remov$4 near3 mercury) and (alkaline OR alkali)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 14:56
S35	46	sorbent and (remov$4 near3 mercury) and (alkaline OR alkali) and (bromine OR iodine)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/25 16:44
S36	15	((bromine OR iodine) same (activated carbon) same react$4) and (remove near3 mercury)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 08:19
S37	1	"6848374" and (alkaline OR alkali)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 09:12
S38	0	"6848374" and (regenerat$)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 09:12
S39	62	((regenerat$4) with sorbent) and (remov$4 near2 mercury)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 09:13
S40	10	((regenerat$4) with sorbent) and (remov$4 near2 mercury) and (bromine OR iodine)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 09:38
S41	0	((regenerat$4) with sorbent) and (remov$4 near2 mercury) and (bromine OR iodine) and regeneragt$4	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 09:39
S42	10	((regenerat$4) with sorbent) and (remov$4 near2 mercury) and (bromine OR iodine) and regenera\t$4	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 09:55

S43	10	((regenerat$4) with sorbent) and (remov$4 near2 mercury) and (bromine OR iodine) and regenerat$4	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 09:39
S44	5	((regenerat$4) with sorbent) and (remov$4 near2 mercury) and (bromine OR iodine) and regenera\t$4 and monitor$3	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 13:42
S45	34	(monitor$3 with mercury) and (remov$4 near2 mercury) and (bromine OR iodine)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 10:00
S46	2	2004/0003716 and monitor$4	US-PGPUB; USPAT	OR	OFF	2007/07/26 10:53
S47	1	"20040003716" and monitor$4	US-PGPUB; USPAT	OR	OFF	2007/07/26 10:55
S48	5	((regenerat$4) with sorbent) and (remov$4 near2 mercury) and (bromine OR iodine) and monitor$3	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 13:42
S49	206	(remov$4 near2 mercury) and (bromine OR iodine) and monitor $3	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 13:42
S50	34	(remov$4 near2 mercury) and (bromine OR iodine) and (monitor $3 with mercury)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 13:53
S51	74	(remov$4 near2 mercury) and (bromine OR iodine) and ((monitor$3 or measur $4) with mercury)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 13:54
S52	38	(remov$4 near2 mercury) and (bromine OR iodine) and ((monitor$3 or measur $4) with mercury) and sorbent	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 14:05
S53	37	(remov$4 near2 mercury) and (bromine OR iodine) and ((monitor$3 or measur $4) with mercury) and sorbent and measur$4	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 15:38

S54	1	(remov$4 near2 mercury) and (bromine OR iodine) and ((monitor$3 or measur$4) with mercury) and sorbent and measur$4 and regenerate	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 14:09
S55	2	(remov$4 near2 mercury) and (bromine OR iodine) and ((monitor$3 or measur$4) with mercury) and sorbent and measur$4 and (particle size near4 "40")	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 15:39
S56	5	(remov$4 near2 mercury) and (bromine OR iodine) and (particle size near4 "40")	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/30 08:21
S57	2	(remov$4 near2 mercury) and (bromine OR iodine) and (particle size near4 "50")	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 15:45
S58	1	(remov$4 near2 mercury) and (bromine OR iodine) and (particle size near4 "60")	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 15:45
S59	2	(remov$4 near2 mercury) and (bromine OR iodine) and (particle size near4 "70")	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 15:47
S60	4	(remov$4 near2 mercury) and (bromine OR iodine) and (particle size near4 "80")	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/26 15:47
S61	5	(remov$4 near2 mercury) and (bromine OR iodine) and (particle size near4 "40") and "40"	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/08/07 16:51
S62	6	(remov$4 near2 mercury) and (bromine OR iodine) and ("50" micrometers)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/30 08:13
S63	1	(remov$4 near2 mercury) and (bromine OR iodine) and ("60" micrometers)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/30 08:14

S65	0	(remov$4 near2 mercury) and (bromine OR iodine) and (separation with (physical means))	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/30 08:23
S67	0	(remov$4 near2 mercury) and (bromine OR iodine) and (separat $4 with (physical means))	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/30 08:23
S68	0	(remov$4 near2 mercury) and (bromine OR iodine) and (remov $4 with (physical means))	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/07/30 08:24
S69	8	(remov$4 near2 mercury) and (bromine OR iodine) and (particle size greater)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/08/07 16:04
S70	430744	alkaline	US-PGPUB; USPAT; USOCR	OR	OFF	2007/08/07 16:02
S71	1	alkaline and "11209163"	US-PGPUB; USPAT; USOCR	OR	OFF	2007/08/07 16:02
S72	1077	(remov$4 near2 mercury) and alkaline	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/08/07 16:04
S73	397	(remov$4 near2 mercury) and alkaline earth	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/08/07 16:04
S74	1	(remov$4 near2 mercury) and (bromine OR iodine) and (separate near3 size)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/08/07 16:51
S75	29	(remov$4 near2 mercury) and (bromine OR iodine) and (separat $4 near3 size)	US-PGPUB; USPAT; USOCR	ADJ	ON	2007/08/07 16:52
S76	13	95/142.ccls. and remov $4 with mercury	US-PGPUB; USPAT; USOCR	OR	OFF	2008/01/16 16:35
S77	1	"11209163"	US-PGPUB; USPAT; USOCR	OR	OFF	2008/01/16 16:35
S78	103	95/134.ccls. and remov $4 with mercury	US-PGPUB; USPAT; USOCR	OR	OFF	2008/01/16 16:35

S79	8	95/134.ccls. and remov $4 with mercury and halide	US-PGPUB; USPAT; USOCR	OR	OFF	2008/01/17 11:12
S80	120	lime and remov$4 with mercury and halide	US-PGPUB; USPAT; USOCR	OR	OFF	2008/01/17 11:13
S82	40	lime and remov$4 with mercury and halide and activated carbon	US-PGPUB; USPAT; USOCR	ADJ	OFF	2008/01/17 11:13
S83	40	lime and remov$4 with mercury and halide and activated carbon	US-PGPUB; USPAT; USOCR	ADJ	OFF	2008/01/17 11:15
S84	76	remov$4 with mercury and lime same activated carbon	US-PGPUB; USPAT; USOCR	ADJ	OFF	2008/01/17 11:19
S85	20	remov$4 with mercury and lime same activated carbon same react$4	US-PGPUB; USPAT; USOCR	ADJ	OFF	2008/01/17 15:03
S86	25	110/345.ccls. and sorbent same mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2008/01/17 15:03
S87	119	423/210.ccls. and (sorbent or bromine or iodine) same mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2008/01/17 15:04
S88	4	active with site and remov$4 with mercury and carbon with halide	US-PGPUB; USPAT; USOCR	OR	OFF	2008/06/30 15:49
S89	7	95/134.ccls. and remov $4 with mercury and carbon with halide	US-PGPUB; USPAT; USOCR	OR	OFF	2008/06/30 15:50
S90	1	95/142.ccls. and remov $4 with mercury and carbon with halide	US-PGPUB; USPAT; USOCR	OR	OFF	2008/06/30 15:56
S91	0	95/142.ccls. and alkaline with inject	US-PGPUB; USPAT; USOCR	OR	OFF	2008/06/30 15:57
S92	0	95/142.ccls. and alkaline with inject$4	US-PGPUB; USPAT; USOCR	OR	OFF	2008/06/30 15:57
S93	1	95/134.ccls. and alkaline with inject$4	US-PGPUB; USPAT; USOCR	OR	OFF	2008/06/30 15:58
S94	151	alkaline with inject$4 and remov$4 with mercury	US-PGPUB; USPAT; USOCR	OR	OFF	2008/06/30 15:58

S95	21	alkaline with inject$4 and remov$4 with mercury and sorbent with inject$4	US-PGPUB; USPAT; USOCR	OR	OFF	2008/06/30 15:59
S96	3	alkaline with inject$4 and remov$4 with mercury and halide with inject$4	US-PGPUB; USPAT; USOCR	OR	OFF	2008/06/30 15:59
S97	0	110/345.ccls. and remov $4 with mercury and carbon with halide	US-PGPUB; USPAT; USOCR	ADJ	OFF	2008/07/01 12:46
S98	5	423/210.ccls. and remov $4 with mercury and carbon with halide	US-PGPUB; USPAT; USOCR	ADJ	OFF	2008/07/01 12:46
S99	0	(halogenated carbon with halide with covalently$5).clm.	US-PGPUB; USPAT; USOCR	ADJ	OFF	2008/07/01 13:05
S100	0	(carbon with haldie with sorbent and reduc $ with mercury).clm.	US-PGPUB; USPAT; USOCR	ADJ	OFF	2008/07/01 13:06
S101	0	(carbon with haldie with sorbent and co-injecting).clm.	US-PGPUB; USPAT; USOCR	ADJ	OFF	2008/07/01 13:07
S102	1	(carbon with sorbent and co-injecting).clm.	US-PGPUB; USPAT; USOCR	ADJ	OFF	2008/07/01 13:07
S103	135	carbon with alkali same remov$4 with mercury	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 16:18
S104	55234	carbon with alkali (salts or elements) same remov$4 with mercury	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 16:18
S105	2	carbon with alkali (salts or elements) same remov$4 with mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:19
S106	135	carbon with alkali same remov$4 with mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:19
S107	15	carbon with alkali same remov$4 with mercury and exhaust	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:19
S108	4	mercury with exhaust and mercury with stabili $5 with reagent and carbon with remov$4 with mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:29

S109	6	"6848374" and (stabili $4 or sulfur or sulfide)	US-PGPUB; USPAT; USOCR	ADJ	ON	2009/04/01 16:31
S110	0	mercury with exhaust and mercury with stabili $5 with (hydrogen sulfide or sulfer dioxide) and carbon with remov$4 with mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:34
S111	0	mercury with exhaust and mercury with stabili $5 with (hydrogen sulfide or sulfur dioxide) and carbon with remov$4 with mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:34
S112	2	stabili$5 with (hydrogen sulfide or sulfur dioxide) and carbon with remov$4 with mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:35
S113	2	mercury with exhaust and mercury with stabili $5 with sulfur and carbon with remov$4 with mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:36
S114	4	"6848374" and (organic or solvent)	US-PGPUB; USPAT; USOCR	ADJ	ON	2009/04/01 16:46
S115	5	"6848374" and (organic or solvent or dissolv$4)	US-PGPUB; USPAT; USOCR	ADJ	ON	2009/04/01 16:46
S116	7	carbon with alkali same remov$4 with mercury and (fixed or moving) bed	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 17:21
S117	8	carbon with halogen same remov$4 with mercury and (fixed or moving) bed	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 17:24
S118	5	carbon with halogen same remov$4 with mercury and (monolith or fiber screens)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 17:30
S119	20	activated carbon same remov$4 with mercury and (monolith or fiber screens)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 17:31

S120	0	activated carbon same remov$4 with mercury and activated carbong with (monolith or fiber screens)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 17:32
S121	2	activated carbon same remov$4 with mercury and activated carbon with (monolith or fiber screens)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 17:32
S122	2	activated carbon same remov$4 with mercury and activated carbon with (monolith or fiber screen)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 17:32
S123	67	"4,889,698"	US-PGPUB; USPAT; USOCR	ADJ	ON	2009/04/01 17:36
S124	125	95/134.ccls. and remov $4 with mercury	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 19:25
S125	20	95/134.ccls. and remov $4 with mercury and halogen	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 19:26
S126	3	95/142.ccls. and remov $4 with mercury and carbon and halide	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 19:26
S127	0	11//345.ccls. and remov $4 with mercury and carbon and (halide or halogen)	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 19:28
S128	8	110/345.ccls. and remov $4 with mercury and carbon and (halide or halogen)	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 19:28
S129	54	423/210.ccls. and remov $4 with mercury and carbon and (halide or halogen)	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 19:28
S130	7	"6848374"	US-PGPUB; USPAT; USOCR	ADJ	ON	2009/04/01 19:34
S131	16	"5575982"	US-PGPUB; USPAT; USOCR	ADJ	ON	2009/04/01 19:35
S133	61	"4500327"	US-PGPUB; USPAT; USOCR	ADJ	ON	2010/01/11 11:30

S135	547	"1984164" "3194629" "3662523" "3786619" "3826618" "3849267" "4101631" "4196173" "4500327" "4708853" "4786483" "4814152" "4820318" "4889698" "4956162" "5300137" "5409522" "5505766" "5575982" "5607496" "5672323" "5695726" "5827352" "5891324" "6027551" "6080281" "6136072" "6214304" "6258334" "6372187" "6375909" "6383981" "6528030" "6818043" "6848374" "6878358" "6942840" "6953494" "7435286" "20010002387" "20010003116" "20020150516" "20040003716" "20040013589" "20050019240" "20060048646"	US-PGPUB; USPAT; USOCR	OR	OFF	2010/01/11 14:38
S136	547	"1984164" "3194,629" "3662,523" "3786619" "3826618" "3849267" "4101631" "4196173" "4500327" "4708853" "4786483" "4814152" "4820318" "4889698" "4956162" "5300137" "5409522" "5505766" "5575982" "5607496" "5672323" "5695726" "5827352" "5891324" "6027551" "6080281" "6136072" "6214304" "6258334" "6372187" "6375909" "6383981" "6528030" "6818043" "6848374" "6878358" "6942840" "6953494" "7435286" "20010002387" "20010003116" "20020150516" "20040003716" "20040013589" "20050019240" "20060048646" and	US-PGPUB; USPAT; USOCR	OR	OFF	2010/01/11 14:47

		Group adj V				
S137	10	("1984164" or "3194629" or "3662523" or "3786619" or "3826618" or "3849267" or "4101631" or "4196173" or "4500327" or "4708853" or "4786483" or "4814152" or "4820318" or "4889698" or "4956162" or "5300137" or "5409522" or "5505766" or "5575982" or "5607496" or "5672323" or "5695726" or "5827352" or "5891324" or "6027551" or "6080281" or "6136072" or "6214304" or "6258334" or "6372187" or "6375909" or "6383981" or "6528030" or "6818043" or "6848374" or "6878358" or "6942840" or "6953494" or "7435286" or "20010002387" or "20010003116" or "20020150516" or "20040003716" or "20040013589" or "20050019240" or "20060048646") and (Group V or Group VI)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2010/01/11 14:50

S138	9	("1984164" or "3194629" or "3662523" or "3786619" or "3826618" or "3849267" or "4101631" or "4196173" or "4500327" or "4708853" or "4786483" or "4814152" or "4820318" or "4889698" or "4956162" or "5300137" or "5409522" or "5505766" or "5575982" or "5607496" or "5672323" or "5695726" or "5827352" or "5891324" or "6027551" or "6080281" or "6136072" or "6214304" or "6258334" or "6372187" or "6375909" or "6383981" or "6528030" or "6818043" or "6848374" or "6878358" or "6942840" or "6953494" or "7435286" or "20010002387" or "20010003116" or "20020150516" or "20040003716" or "20040013589" or "20050019240" or "20060048646") and (Group V or Group VI) and (Hg or mercury)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2010/01/11 14:52
S139	46	(Group V or Group VI) with (halide or halogen) and (Hg or mercury) with (filter or remov$4)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2010/01/11 14:53

S141	68	95/134.ccls. and remov $4 with mercury and (V or Nb or Ta or Db or Cr or Mo or W or Sg or Group V or Group VI)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2010/01/11 14:57
S142	10	110/345.ccls. and remov $4 with mercury and carbon and (halide or halogen)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2010/01/11 15:08
S144	64	423/210.ccls. and remov $4 with mercury and carbon and (halide or halogen)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2010/01/11 15:09
S145	267	(Vanadium or niobium or tantalum or Dubnium or chromium or molybdenum or tungsten or seaborgium) with halide and (remov$4 or filter) with (mercury or Hg)	US-PGPUB; USPAT; USOCR	ADJ	ON	2010/01/12 13:08
S146	58	(Vanadium or niobium or tantalum or Dubnium or chromium or molybdenum or tungsten or seaborgium) with sorbent and (remov$4 or filter) with (mercury or Hg)	US-PGPUB; USPAT; USOCR	ADJ	ON	2010/01/12 13:26
S147	4	(Vanadium or niobium or tantalum or Dubnium or chromium or molybdenum or tungsten or seaborgium) with halide same sorbent and (remov$4 or filter) with (mercury or Hg)	US-PGPUB; USPAT; USOCR	ADJ	ON	2010/01/12 13:28
S148	4	(Vanadium or niobium or tantalum or Dubnium or chromium or molybdenum or tungsten or seaborgium) with (halide or halogen) same sorbent and (remov$4 or filter) with (mercury or Hg)	US-PGPUB; USPAT; USOCR	ADJ	ON	2010/01/12 13:28

S149	8287	hydrohalides	US-PGPUB; USPAT; USOCR	ADJ	ON	2010/01/12 13:45
S150	5	hydrohalides with sorbent	US-PGPUB; USPAT; USOCR	ADJ	ON	2010/01/12 13:45
S151	143	hydrohalides.ti.	US-PGPUB; USPAT; USOCR	ADJ	ON	2010/01/12 13:46
S152	5	"4490354"	US-PGPUB; USPAT; USOCR	ADJ	ON	2010/01/12 13:59
S153	10	"6,136,749"	US-PGPUB; USPAT; USOCR	OR	OFF	2010/01/12 14:34

EAST Search History (Interference)

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2/ 26/ 2010 3:16:25 PM
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Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	12201595	OLSON ET AL.
	Examiner	**Art Unit**
	AMBER ORLANDO	1797

SEARCHED

Class	Subclass	Date	Examiner
110	345	4/1/2009	AO
423	210	4/1/2009	AO
95	134, 142	4/1/2009	AO
110	345	2/26/2010	AO
423	210	2/25/2010	AO
95	134, 142	2/26/2010	AO

SEARCH NOTES

Search Notes	Date	Examiner
Inventor Search (eDAN)	4/1/2009	AO
Class 95 searched for a carbon sorbent having a promoter. (EAST)	4/1/2009	AO
Classes 423 and 110 searched for a carbon sorbent having a halide promoter and an alkaline component. (EAST)	4/1/2009	AO
Classes 95, 423 and 110 searched for new art (EAST)	2/26/2010	AO

INTERFERENCE SEARCH

Class	Subclass	Date	Examiner

/AMBER ORLANDO/ Acting Examiner.Art Unit 1797	

Index of Claims	Application/Control No.	Applicant(s)/Patent Under Reexamination
	12201595	OLSON ET AL.
	Examiner	Art Unit
	AMBER ORLANDO	1797

✓	Rejected	-	Cancelled	N	Non-Elected	A	Appeal
=	Allowed	÷	Restricted	I	Interference	O	Objected

☐ Claims renumbered in the same order as presented by applicant ☐ CPA ☐ T.D. ☐ R.1.47

CLAIM		DATE								
Final	Original	04/01/2009	04/01/2009	02/26/2010						
	1	✓		✓						
	2	✓		✓						
	3	✓		✓						
	4	✓		-						
	5	✓		-						
	6	✓		-						
	7	✓		✓						
	8	✓		-						
	9	✓		-						
	10	✓		✓						
	11	✓		-						
	12	✓		-						
	13	✓		-						
	14	✓		✓						
	15	✓		✓						
	16	✓		✓						
	17	✓		-						
	18	✓		-						
	19	✓		-						
	20	✓		-						
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	27	✓		-						
	28	✓		-						
	29	✓		-						
	30	✓		-						
	31	✓		-						
	32	✓		-						
	33	✓		-						
	34			✓						
	35			✓						

FORM 1449* INFORMATION DISCLOSURE STATEMENT IN AN APPLICATION (Use several sheets if necessary)	Docket Number: 16291.0003USD1	Application Number: 12/201,595
	Applicant: Olson et al.	
	Filing Date: 08/29/2008	Group Art Unit: 1797

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	4,101,631	07-18-1978	Ambrsini, et al.			
	4,196,173	04-01-1980	Delong, et al.			
	4,500,327	02-19-198	Nishino, et al.			
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	5,607,496	03-04-1997	Brooks			
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	5,695,726	12-09-1997	Lerner			
	5,827,352	12-27-1998	Altman			
	5,891,324	04-06-1999	Ohtsuka			
	6,027,551	02-22-2000	Hwang			
	6,080,281	06/27/2000	Attia			
	6,136,072	10/24/2000	Sjostrom			
	6,214,304	04-10-2001	Rosenthal			
	6,258,334	07-10-2001	Gadkaree			
	6,383,981	05-07-2002	Blankenship			

EXAMINER	/Amber Orlando/	DATE CONSIDERED	02/26/2010

EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609; draw line through citation if not in conformance and not considered. Include copy of this form for next communication to the Applicant.

*Substitute Disclosure Statement Form (PTO-1449) Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

ALL REFERENCES CONSIDERED EXCEPT WHERE LINED THROUGH. /A.O./

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	6,953,494	10-11-2005	Nelson, Jr.			
	7,435,286	10-14-2008	Olson et al.			
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	2004/0003716	01-08-2004	Nelson, Jr.			
	2004/0013589	01-22-2004	Vosteen et al.			
	2005/0019240	01-2005	Lu et al.			

FOREIGN PATENT DOCUMENTS

	DOCUMENT NO.	DATE	COUNTRY	CLASS	SUBCLASS	TRANSLATION	
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	2150529	12/29/1995	Canada				
	3426059	01/1986	Germany				
	10233173	03/23/2006	Germany				
	0208490A1	01/1987	Europe				
	49-43197	11/1974	Japan				
	49-53593	05/24/1974	Japan			Abstract	
	50-6438	03/1975	Japan				
	01/62368	08/30/2001	PCT				

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OTHER DOCUMENTS (Including Author, Title, Date, Pertinent Pages, Etc.)		
		Dunham, G.E. et al., *Mercury Capture by an Activated Carbon in a Fixed-Bed Bench-Scale System*, Environmental Progress Vol. 17, No. 3, PP. 203-208
		Felsvang, K. et al., *Mercury Reduction and Control Options,* The U.S. EPA-DOE-EORI Combined Power Plant Air Pollutant Control Symposium: The Mega Symposium and The A&WMA Specialty Conference on Mercury Emissions: Fate, Effects and Control. August 20-23, 2001, Chicago, Illinois, pp. 1-19
		Ghorishi, B; Guller, B.K., *Sorption of Mercury Species by Activated Carbons and Calcium-Bases Sorbents: Effect of Temperature, Mercury Concenitration and Acid Gases, Waste Manage.* Res. 1998, 16, 6 582-593.
		Lancia, A.; Musmarra, D.; Pepe, F.; Volpicelli, G., *Adsorption of Mercuric Chloride Vapours from Incinerator Flue Gases on Calcium Hydroxide Particles,* Combust. Sci. & Technol. 1993, 93, 277.
		Olson, D.G.; et al., *The Reduction of Gas Phase Air Toxics from Combustion and Incineration Sources using the MET-Mitsui-BF Activated Coke Process,* Fuel Processing Technology, 65-66 (2000) 393-405
		B. Vosteen et al.,*Chlor-und Bromgestützte Hg-Sorption an Elektrofilter-Flugaschen eines kohlegefeuerten Schmelzkammerkessels und an Zementrohmehl,* Matin-Luther-Universität Halle-Wittenberg, September 2003, pp 1-30

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PATENT TRADEMARK OFFICE

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant:	OLSON ET AL.	Examiner:	A. ORLANDO
Serial No.:	12/201,595	Group Art Unit:	1797
Filed:	AUGUST 29, 2008	Docket No.:	16291.3USD1
Title:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY		

Electronically filed on December 1, 2009

INFORMATION DISCLOSURE STATEMENT (37 C.F.R. § 1.97(c))

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

Dear Sir:

With regard to the above-identified application, the items of information listed on the enclosed Form 1449 are brought to the attention of the Examiner.

This statement should be considered because it is submitted after the mailing date of a first Office Action on-the-merits or a first Office Action after filing a Request for Continued Examination under 37 C.F.R. § 1.114 or a CPA under 37 C.F.R. § 1.53(d), but before the mailing date of: i) a final action under 37 C.F.R. § 1.113; ii) a Notice of Allowance under 37 C.F.R. § 1.311; or iii) an action that otherwise closes prosecution on the application. Payment is being made via credit card in the amount of $180.00 under 37 C.F.R. § 1.17(p) for consideration of the items listed on the enclosed Form 1449.

In accordance with 37 C.F.R. §1.98(a)(2), a copy of each foreign patent and/or a copy of each publication, other than U.S. patents and U.S. patent application publications, listed on the accompanying Form 1449 is enclosed.

No representation is made that a reference is "prior art" within the meaning of 35 U.S.C. §§ 102 and 103 and Applicants reserve the right, pursuant to 37 C.F.R. § 1.131 or otherwise, to establish that the reference(s) are not "prior art." Moreover, Applicants do not represent that a reference has been thoroughly reviewed or that any relevance of any portion of a reference is intended.

Enclosed for the Examiner's information, as Attachment A, is a copy of a Communication from the European Patent Office dated January 25, 2008 and a copy of the "Third party's comments following 115 EPO about the patent applications EP 1796816A2 and related PTC patent applications as WO 2006/039007 of EERC" that is the subject of the Communication. European patent application EP 1796816A2 and PCT publication WO 2006/039007 (PCT Application PCT/US05/030018) are related to the present application.

Also enclosed for the Examiner's information, as Attachment B, is a copy of the European Search Report for EP1796816, dated 14 April 2009. For the Examiner's reference, all citations within the European Search Report are identified on the accompanying Form 1449.

The Examiner's attention is directed to co-pending United States Patent Application Serial Nos. 12/429,058 and 12/419,219. These applications are related to the present application.

Consideration of the items listed is respectfully requested. Pursuant to the provisions of M.P.E.P. 609, it is requested that the Examiner return a copy of the attached Form 1449, marked as being considered and initialed by the Examiner, to the undersigned with the next official communication.

Please charge any additional fees or credit any overpayment to Deposit Account No. 13-2725.

Respectfully submitted,

MERCHANT & GOULD P.C.
P.O. Box 2903
Minneapolis, Minnesota 55402-0903
(612) 332-5300

Date: November 10, 2009

Dennis R. Daley
Reg. No. 34,994
DDaley:TPJ:rlk

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PATENT TRADEMARK OFFICE

FORM 1449* INFORMATION DISCLOSURE STATEMENT IN AN APPLICATION (Use several sheets if necessary)	Docket Number: 16291.0003USD1	Application Number: 12/201,595
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	5,607,496	03-04-1997	Brooks			
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	2004/0013589	01-22-2004	Vosteen et al.			
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		Ghorishi, B; Guller, B.K., *Sorption of Mercury Species by Activated Carbons and Calcium-Bases Sorbents: Effect of Temperature, Mercury Concenitration and Acid Gases, Waste Manage.* Res. 1998, 16, 6 582-593.
		Lancia, A.; Musmarra, D.; Pepe, F.; Volpicelli, G., *Adsorption of Mercuric Chloride Vapours from Incinerator Flue Gases on Calcium Hydroxide Particles,* Combust. Sci. & Technol. 1993, 93, 277.
		Olson, D.G.; et al., *The Reduction of Gas Phase Air Toxics from Combustion and Incineration Sources using the MET-Mitsui-BF Activated Coke Process,* Fuel Processing Technology, 65-66 (2000) 393-405
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OPIC
Office de la propriété intellectuelle du Canada

CIPO
Canadian Intellectual Property Office

Ottawa Hull K1A 0C9

(21) (A1) 2,150,529
(22) 1995/05/30
(43) 1995/12/29

(51) Int.Cl.[6] B01D 53/64

(19)(CA) **APPLICATION FOR CANADIAN PATENT** (12)

(54) Method and Apparatus for Continuously Separating Mercury from Flowing Gases

(72) Scherer, Volker - Germany (Federal Republic of) ;
Greczmiel, Monika - Germany (Federal Republic of) ;

(71) Patent-Treuhand-Gesellschaft für elektrische Glühlampen mbH - Germany (Federal Republic of) ;

(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

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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.

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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.

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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.

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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.

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

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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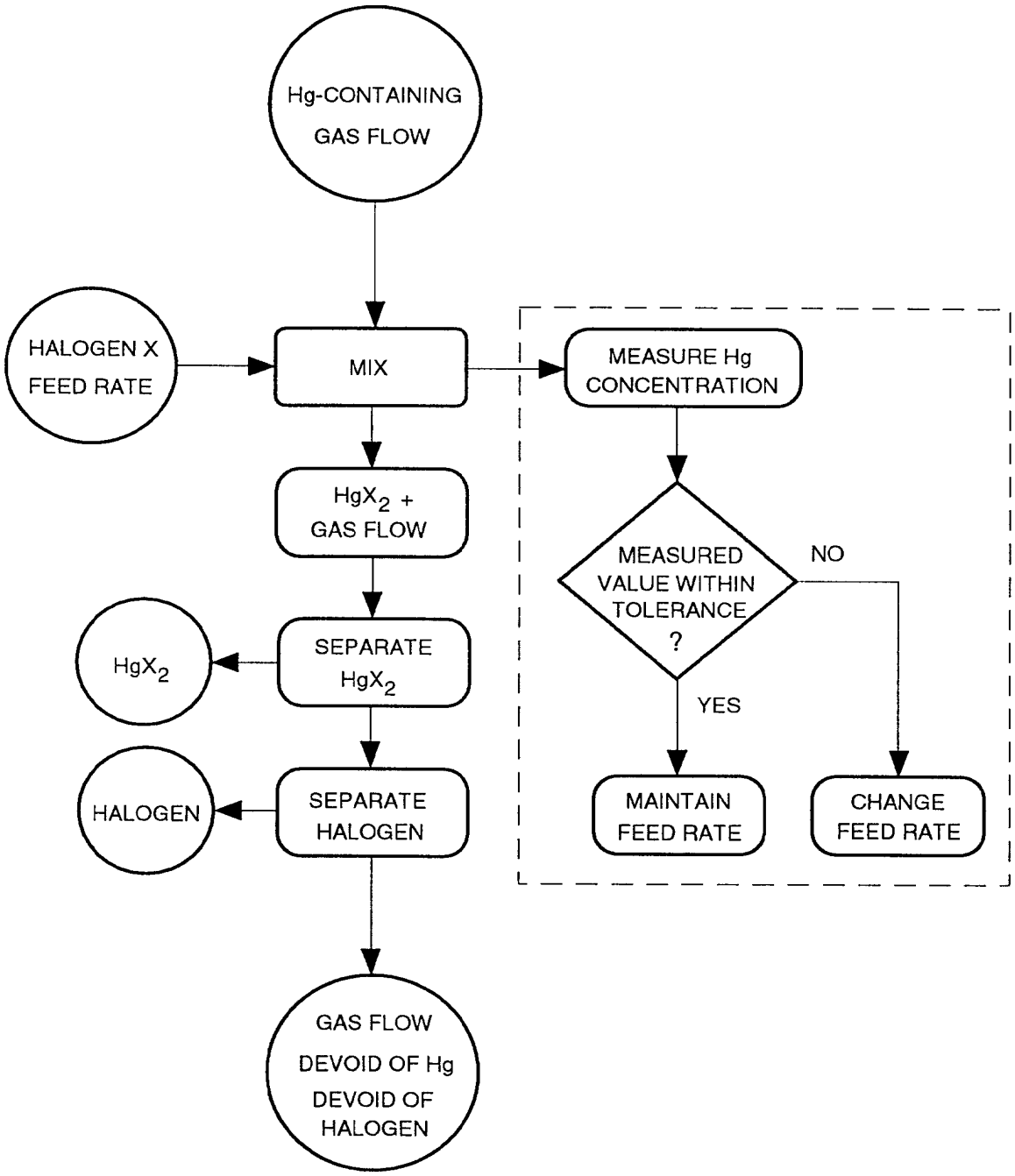


FIG. 1

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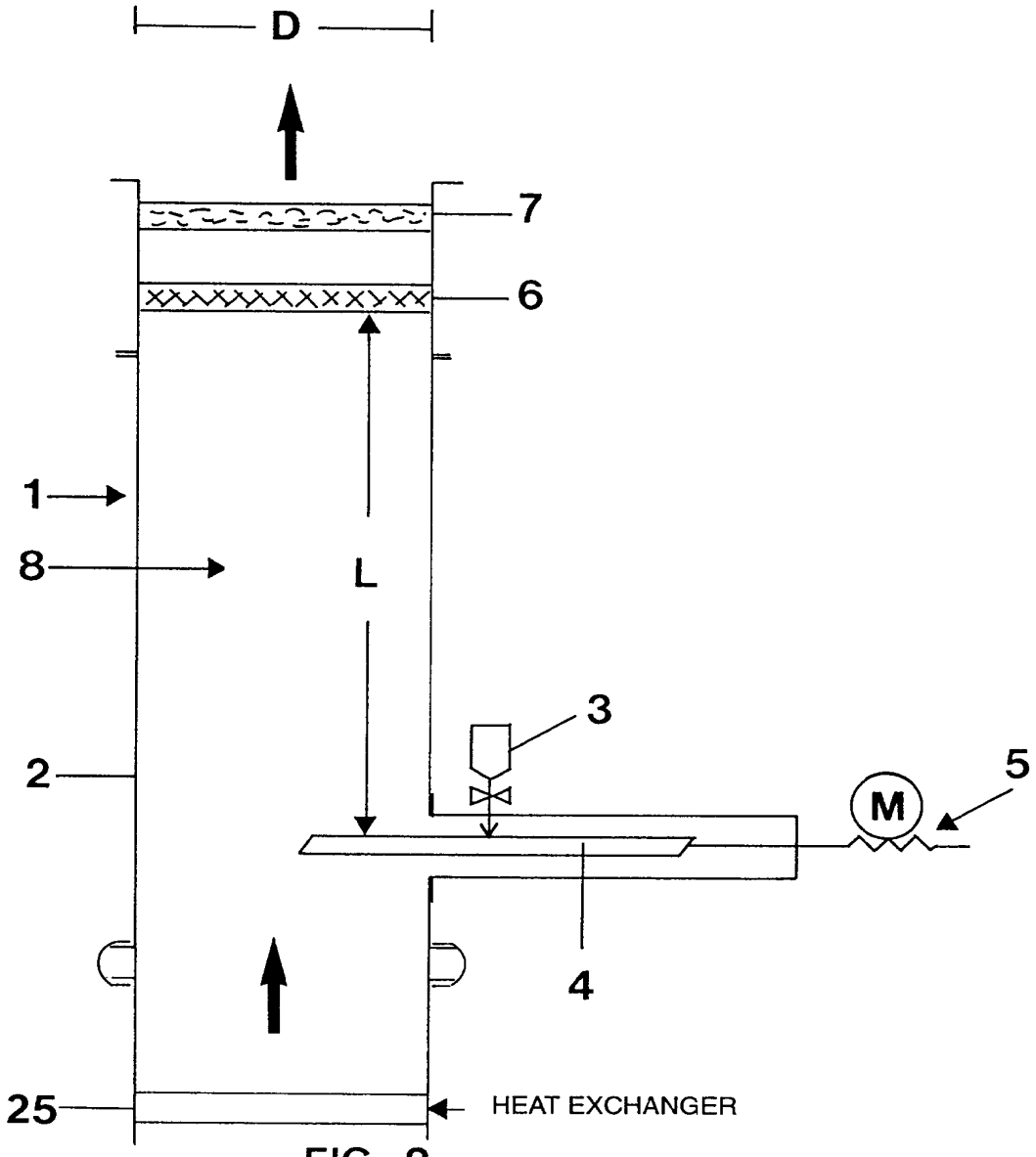


FIG. 2

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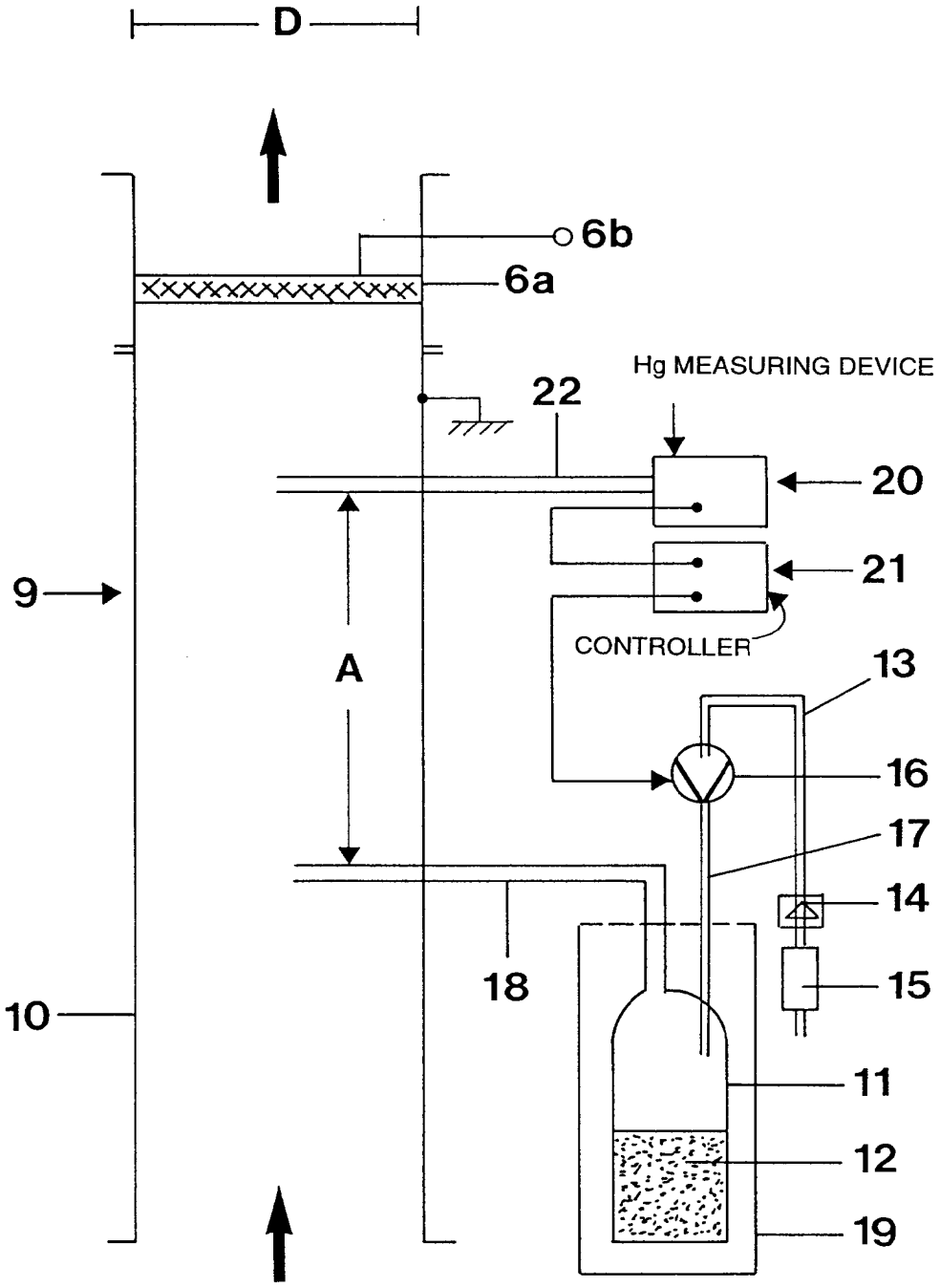


FIG. 3

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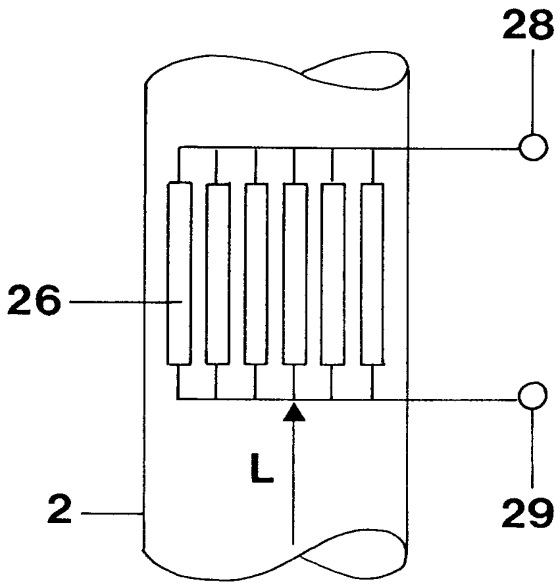


FIG. 4

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(54) Verfahren zur Abscheidung und Beseitigung von organischen Schadstoffen, insbesondere von polychlorierten Dibenzodioxinen (PCDD) aus Rauchgasen

Die Erfindung betrifft ein Verfahren zur Abscheidung und Beseitigung von organischen Schadstoffen, insbesondere polyhalogenierten, aromatischen Verbindungen aus Rauchgasen oder sonstigen Abgasen durch thermische Zersetzung der Schadstoffe bei Temperaturen von über 1000 °C, wobei die organischen Schadstoffe aus dem Rauchgas zunächst adsorptiv, vorzugsweise mittels eines Aktivkohlefilters, entfernt, danach bei erhöhten Temperaturen desorbiert und aus dem Desorptionsgas, das nur einen Bruchteil des Volumens des Rauchgases ausmacht, durch Verbrennung bei Temperaturen von 1000 bis 1400 °C entfernt werden. Die Aufheizung des Desorptionsgases erfolgt mittels Fremdwärme. Zweckmäßig wird der Wärmeinhalt des heißen Verbrennungsgases, beispielsweise zur Vorwärmung der Verbrennungsluft ausgenutzt.

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Patentansprüche

1. Verfahren zur Abscheidung und Beseitigung von schwer zersetzlichem organischen Schadstoffen, insbesondere polyhalogenierten aromatischen Verbindungen aus Rauchgasen durch Erhitzen des schadstoffhaltigen Rauchgases auf Temperaturen von > 1000°C, dadurch gekennzeichnet, dass die Schadstoffe aus dem Rauchgas adsorptiv entfernt, die adsorptiv gebundenen Schadstoffe durch Erhitzen auf höhere Temperaturen desorbiert und das Desorptionsgas durch Zuführung von Fremdwärme auf Temperaturen von über 1000°C, vorzugsweise 1000 bis 1400°C bei einer zur Spaltung der Schadstoffe ausreichenden Verweilzeit erhitzt werden.

2. Verfahren nach Anspruch 1, dadurch gekennzeichnet, dass die organischen Schadstoffe durch Adsorption mittels Aktivkoks oder Aktivkohle aus dem Rauchgas entfernt werden.

3. Verfahren nach einem der Ansprüche 1 oder 2, dadurch gekennzeichnet, dass die Desorption der organischen Schadstoffe bei Temperaturen im Bereich von 350 bis 750° zusammen mit anderen absorbiereten Schadstoffen vorgenommen wird.

4. Verfahren nach einem der Ansprüche 1 bis 3, dadurch gekennzeichnet, dass die thermische Spaltung der Schadstoffe im Desorptionsgas in einer Spaltkammer erfolgt.

5. Verfahren nach einem der Ansprüche 1 oder 2, dadurch gekennzeichnet, dass die Desorption und die thermische Zersetzung der Schadstoffe mittels des auf die Zersetzungstemperaturen aufgeheizten Desorptionsgases vorgenommen wird.

6. Verfahren nach einem der Ansprüche 1 bis 5, dadurch gekennzeichnet, dass das aus der thermischen Zersetzung der Schadstoffe resultierende heiße Abgas unter Ausnutzung seines Wärmeinhalts, vorzugsweise zur Vorwärmung der Verbrennungsluft anderer Gase, oder zur Nutzung im Wasserdampfkreislauf abgekühlt wird.

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Verfahren zur Abscheidung und Beseitigung von organischen Schadstoffen, insbesondere von polychlorierten Dibenzodioxinen (PCDD) aus Rauchgasen

Die Erfindung betrifft ein trockenes Verfahren zur Abscheidung von in Rauchgasen oder in sonstigen Abgasen enthaltenen organischen schwer zersetzlichen Schadstoffen, insbesondere von polychlorierten Dibenzodioxinen.

Bei der Verbrennung von Industrie- und Hausmüll in Müllverbrennungsanlagen entstehen verschiedene organische Schadstoffe, vor allem aromatischer Natur wie

Polyzyklische aromatische Kohlenwasserstoffe (PAK)

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polychlorierte Biphenyle (BCB)
polychlorierte Dibenzodioxine (PCDD)
polychlorierte Dibenzofurane (PCDF)

Wegen der bekannten gesundheitsschädlichen Wirkung ist die Beseitigung dieser Substanzen deshalb von besonderem Interesse, da sie nur schwer abbaubar sind und deswegen die Gefahr einer möglichen Anreicherung in der Natur besteht.

Die Beseitigung der organischen Schadstoffe ist durch Zerstörung der Molekülstrukturen bei hohen Spalttemperaturen meist im Bereich von über 1000°C möglich. Hierbei spielen Spalttemperaturen und die Verweilzeit der organischen Schadstoffe im hohen Temperaturbereich, die meist einige Sekunden betragen soll, eine wesentliche Rolle.

Aus diesem Grund werden Sonderabfälle, vor allem Abfälle aus der chemischen Produktion, vor allem in Sondermüll-Verbrennungsanlagen, die mit Verbrennungs-Reaktionstemperaturen von über etwa 1100°C arbeiten, bei Verweilzeiten der Rauch-/Abgase in diesem hohen Temperaturbereich von bis zu 6 Sekunden verbrannt.

Bau und Betrieb solcher Abfallverbrennungsanlagen sind im Vergleich zu normalen Haus/Industrie-Müllverbrennungsanlagen sehr kostenaufwendig und wirtschaftlich nur dann vertretbar, wenn diese Anlage speziell für die Spaltung dieser organischen Schadstoffe eingesetzt wird.

In normalen Haus-/Industrie-Müllverbrennungsanlagen treten diese Schadstoffe jedoch nicht ständig, sondern

nur dann auf, wenn diese organischen Schadstoffe im zu verbrennenden Abfall anfallen. Die Konzentrationen im Rauchgas sind damit sehr unterschiedlich und können stark schwanken, z.B von einigen Pikogramm bis zu mehreren Hundert Pikogramm .
Eine generelle Auslegung, Bau und Betrieb aller Müll- und Abfallverbrennungsanlagen bei diesen hohen Temperaturen führt neben hohen Kosten für Investition und Betrieb vor allem auch zu anderen Nachteilen, wie beispielsweise höher NO_x-Konzentrationen im Rauchgas und zu erhöhter Korrosionsgefahr an den Verbrennungseinrichtungen, wie beispielsweise den Rosten sowie den Dampfkessel oder Wärmeaustauschern.

Diese Nachteile treten dann nicht auf, wenn es gelingt, diese organischen Schadstoffe aus den Rauchabgasen abzuscheiden und dann in geeigneter Form zu beseitigen.

Gegenstand der Erfindung ist ein Verfahren zur Abscheidung und Beseitigung von schwer zersetzlichen, organischen Schadstoffen, insbesondere polyhalogenierten aromatischen Verbindungen aus Rauchgasen durch Erhitzen des schadstoffhaltigen Rauchgases auf Temperaturen von $>$ 1000°C, dadurch gekennzeichnet, dass die Schadstoffe aus dem Rauchgas adsorptiv entfernt, die adsorptiv gebundenen Schadstoffe durch Erhitzen auf höhere Temperatur desorbiert und das Desorptionsgas durch Zuführung von Fremdwärme auf Temperaturen von über 1000°C, vorzugsweise 1000 bis 1400°C bei einer zur Spaltung der Schadstoffe ausreichenden Verweilzeit erhitzt werden.

Als Adsorptionsmittel sind besonders A-Kohle oder A-Koks geeignet. Dieser A-Koks, A-Kohle oder ein anderes geeignetes Adsorptionsmittel werden in an sich bekannter Weise durch Erhitzen auf höhere Temperaturen desorbiert.

Die Kohleschicht weist eine Dicke von z.B. 1,50 bis 2 m auf.

Da bei der Reinigung von Rauchgasen mittels eines A-Koks (A-Kohle-)Filterbettes auch andere Schadstoffe wie SO_2 und Schwermetalle simultan abgeschieden werden, ist es bisher bekannt, die Desorption dieser A-Kokse/ A-Kohle mit inerten Gasen im Temperaturbereich von etwa 350-750°C durchzuführen. Diese Temperatur reicht damit aber zur thermischen Spaltung der organischen Schadstoffe noch nicht aus.

Gemäß einer Ausführungsform der Erfindung wird das Desorptionsmittel hinter dem Desorber durch Zuführen von Fremdwärme, wie beispielsweise durch Zubrennen von Erdgas, Heizöl, sonstigen fossilen Brennstoffen oder auch durch Erwärmung mit elektrischer Energie in einer Spaltkammer auf Temperaturen von über 1000°C bis zu etwa 1400°C erhitzt. Die Verweilzeit der Gase in der Spaltkammer wird so bemessen, dass genügend Zeit zur vollständigen Spaltung der organischen Schadstoffmoleküle besteht. Sie liegt in der Größenordnung von mehreren Sekunden (z.B. 5 bis 10 Sekunden). Das heiße Gas wird nach der Spaltung wieder mittels eines Wärmeaustauschers abgekühlt, wobei die freiwerdende Wärme in wärmewirtschaftlich sinnvoller Weise, also beispielsweise zur Vorwärmung der Verbrennungsluft anderer Gase, aber auch zur wärmewirtschaftlichen Nutzung im Wasserdampfkreislauf einer solchen Müllverbrennungsanlage ausgenutzt.

Gemäß einer weiteren Ausführungsform der Erfindung kann die Spaltkammer für die thermische Zersetzung der Schadstoffe in Desorptionsgas entfallen. In diesem Falle wird die Desorption gleich mit einem heißen Desorptionsgas vorgenommen, das die zur thermischen Spaltung der Schadstoffe notwendige Wärmeenergie enthält. Diese Ausführungsweise ist möglich, da die A-Kohle des Filters hohe Tempe-

raturen im Bereich von 1000 bis 1300 °C ohne weiteres aushält. Das Desorptionsgas (üblicherweise entweder durch Verbrennung von Heizöl oder Erdgas mit unterstöchometrischen Sauerstoffmengen erhalten) wird dann bei den für die Zersetzung erforderlichen Temperaturen in der Größenordnung von 1200°C in den Desorber geleitet.

Der Vorteil dieses Verfahrens liegt somit darin, dass die in sehr niedrigen Konzentrationen im großen Rauchgasvolumen enthaltenen Schadstoffe zunächst aus diesem herausgefiltert werden und dann lediglich in der sehr viel geringeren Desorptionsmenge, die nun natürlich eine höhere Schadstoffkonzentration aufweist, auf die erforderlichen Spalttemperaturen zu erhitzen, wodurch ein wesentlich wärmewirtschaftlicherer Betrieb erreicht wird, da die Rauchgasmenge um ein vielfaches, teilweise bis zum Faktor 100-200, größer ist als die Desorptionsgasmenge.

Die Erfindung wird anhand der nachfolgenden Zeichnung beispielhaft erläutert:

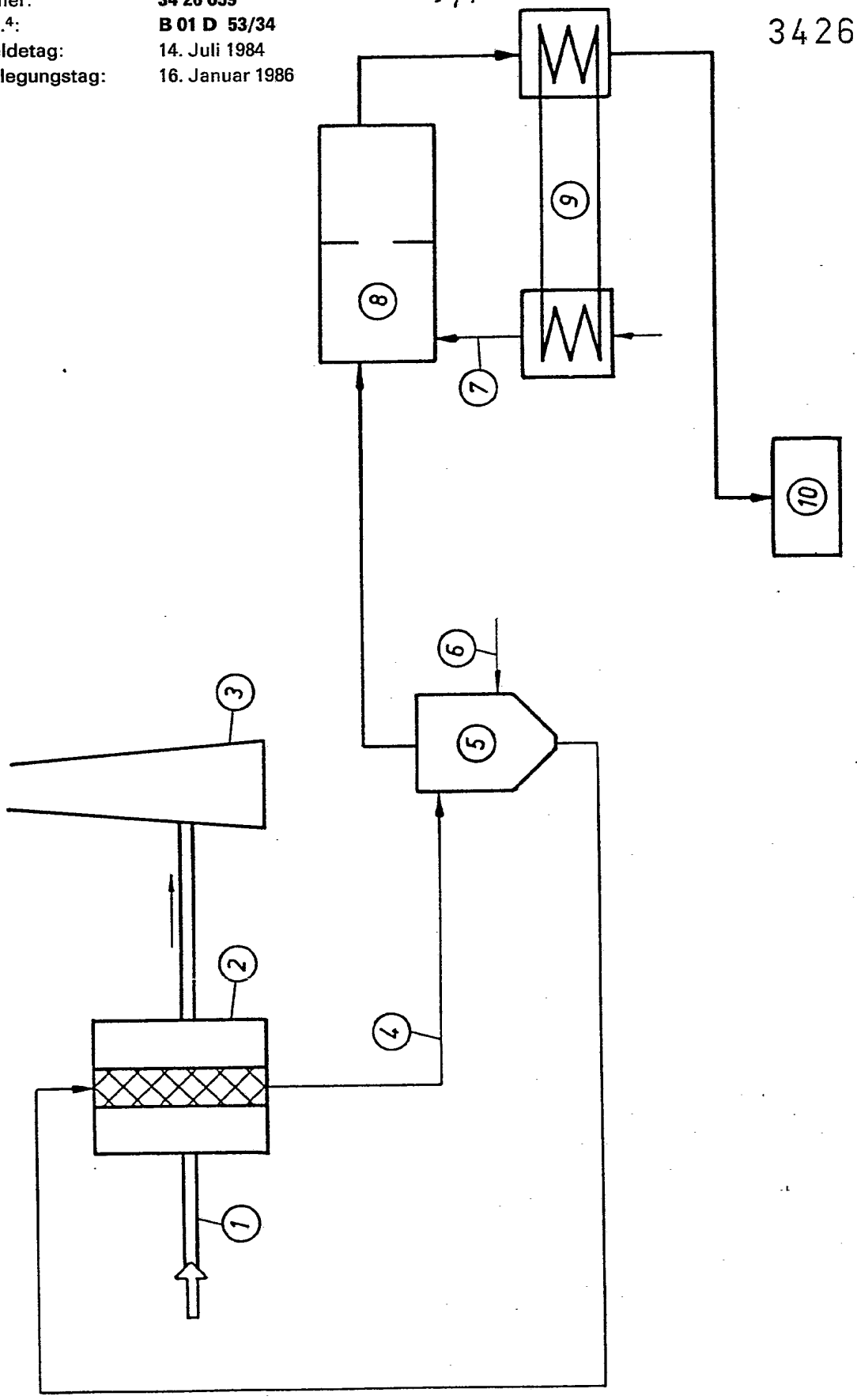
Das mit den organischen Schadstoffen beladene Rauchgas (1) tritt in den Adsorber (2) ein, der mit einem geeigneten Adsorptionsmittel wie Aktivkoks beschickt ist und wird dann über den Schornstein (3) abgeführt. Das mit den Schadstoffen beladene Adsorptionsmaterial wird über Leitung (4) in den Desorber (5) transportiert, dort bei erhöhten Temperaturen mittels eines Desorptionsgases der Schadstoff ausgetrieben und in eine Verbrennungskammer (8) transportiert. Diese wird mittels des durch Leitung (7) zugeführten heißen Gases auf die zur Zersetzung der Schadstoffe erforderlichen Temperaturen von

> 1000°C bis zu etwa 1400°C aufgeheizt. In dieser Spaltkammer erfolgt die thermische Zersetzung bzw. Verbrennung der Schadstoffe. Das heiße Gas verlässt die Spaltkammer und gibt in einem Wärmetauscher (9) wenigstens einen Teil seines Wärmeinhalts an das zur Aufheizung der Kammer (8) verwendete Gas ab, bevor es in die Reinigungsanlage (10) eintritt.

Bei der weiteren Ausführungsform gemäß der Erfindung entfällt die Spaltkammer (8). Bei (6) wird in den Absorber heißes Desorptionsgas eingeleitet, das dann wieder im Wärmeaustauscher (9) wenigstens einen Teil seines Wärmeinhalts abgibt, vorzugsweise zum Aufheizen des gleichzeitig zur Desorption und der thermischen Zersetzung verwendeten in den Desorber eintretenden Gases.

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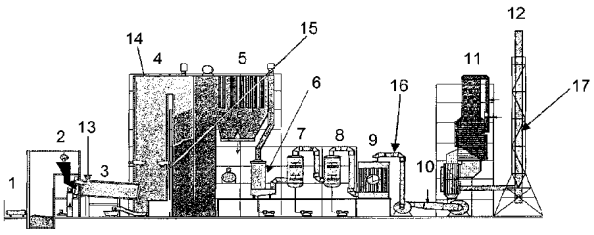
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Prüfungsantrag gemäß § 44 PatG ist gestellt.

Die folgenden Angaben sind den vom Anmelder eingereichten Unterlagen entnommen

(54) Bezeichnung: **Verfahren zur Abscheidung von Quecksilber aus Rauchgasen**

(57) Zusammenfassung: Die Erfindung beschreibt ein Verfahren zur Abscheidung von Quecksilber aus Rauchgasen von Hochtemperaturanlagen, insbesondere von Kraftwerken und Abfallverbrennungsanlagen, bei dem der, gegebenenfalls mehrstufigen, Feuerung und/oder dem Rauchgas in einem der Feuerung nachgeschalteten Anlagenteil eine bromhaltige Verbindung zugeführt wird, wobei die Temperatur beim Kontakt der bromhaltigen Verbindung mit dem Rauchgas mindestens 500°C, vorzugsweise mindestens 800°C, beträgt. Die Verbrennung erfolgt in Gegenwart einer schwefelhaltigen Verbindung, insbesondere Schwefeldioxid. Im Anschluss an die Feuerung wird das Rauchgas einer, gegebenenfalls mehrstufigen, Reinigung zur Abtrennung von Quecksilber aus dem Rauchgas unterzogen, welche eine Nasswäsche und/oder eine Trockenreinigung umfasst.



Beschreibung

[0001] Die Erfindung betrifft ein Verfahren zur Abscheidung von Quecksilber aus Rauchgasen von Hochtemperaturanlagen, insbesondere von Kraftwerken und Abfallverbrennungsanlagen.

Stand der Technik

[0002] Aufgrund der hohen Toxizität von Quecksilber, insbesondere von organisch gebundenem Quecksilber, das direkt oder indirekt über die Nahrungskette auch vom Menschen aufgenommen wird, existieren strenge Grenzwerte für die gesetzlich zulässige Emission von Quecksilber z.B. aus Verbrennungsanlagen und Kraftwerken. Trotz der heute bereits niedrigen Quecksilber-Reingaskonzentrationen – der derzeit in Deutschland zulässige Halbstundenmittelwert für Quecksilberemissionen aus Abfallverbrennungsanlagen beträgt 30 $\mu g/m^3$ i. N. tr. (i. N. tr.: im Normalzustand trocken) – werden aufgrund großer Volumenströme, beispielsweise aus Großkraftwerken, beachtliche Quecksilberfrachten erzielt, so dass eine weitere Minderung der derzeit zulässigen Grenzwerte angestrebt wird.

[0003] Diverse Verfahren zur Minderung von Quecksilberemissionen aus Kraftwerken, Abfallverbrennungsanlagen oder dgl. sind aus der Literatur bekannt. Welches der Verfahren für eine bestimmte Anwendung zweckmäßig ist, hängt stark von der eingebrachten Fracht und vom Chlorgehalt des Feuerungsgutes ab. Bei hohem Chlorgehalt ist der Anteil an ionischem Quecksilber im Rauchgas hoch. Ionisches Quecksilber lässt sich gut in Wäschern abscheiden. Das quasi-wasserunlösliche metallische Quecksilber kann beispielsweise durch den Zusatz von Oxidationsmitteln, wie Peroxide, Ozon oder Natriumchlorit, im Kesselrohgas vor der Rauchgasreinigung oder im entstaubten Kesselrohgas in ionisches Quecksilber überführt und anschließend in Wäschern abgeschieden werden. Weitere Verfahren zur Abscheidung des Quecksilbers sind: der Zusatz von Reaktionsmitteln, wie Natriumtetrasulfit, zur Bindung des Quecksilbers mittels Schwefel im Kesselrohgas vor Rauchgasreinigung oder im teilgereinigten Reingas; die verbesserte Auswaschung von ionischem Quecksilber durch pH- bzw. pCl-Absenkung in der sauren Wäsche oder durch Beaufschlagung mit 1,3,5-Triazin-2,4,6-trithiol (Trimercapto-S-triazin, TMT) in der schwach sauren oder schwach alkalischen Wäsche; die sorptive Abscheidung von ionischem und metallischem Quecksilber unter Zusatz von pulvrigen Sorbentien oder zerstäubten Suspensionen.

[0004] Bisherige Techniken zur Minderung sind nicht ausreichend effektiv und sind aufgrund ihrer z.T. hohen Zusatzinvestitionen und dem zusätzlichen Betriebsmittelverbrauch relativ teuer.

Aufgabenstellung

[0005] Aufgabe der Erfindung ist es, ein Verfahren zur Abscheidung von Quecksilber, insbesondere zur weitgehend vollständigen Abscheidung von Quecksilber (Hg), aus Rauchgasen von Hochtemperaturprozessen bereitzustellen. Das Verfahren soll breitestmögliche Anwendung finden, wie im Falle von im Wesentlichen konstanten niedrigen Hg-Konzentrationen, z.B. in Kohlekraftwerken, aber auch im Falle von relativ hohen Hg-Konzentrationen, z.B. in der Klärschlammverbrennung, oder sehr hohen Hg-Konzentrationen z.B. in der Hausmüll- oder Sondermüllverbrennung. Des Weiteren sollte das Verfahren keiner umfangreichen Nachrüstung der Hochtemperaturanlagen bedürfen sowie möglichst geringer zusätzlicher Betriebsmittel, so dass das Verfahren kostengünstig implementiert und betrieben werden kann.

[0006] Gegenstand der Erfindung ist ein Verfahren zur Abscheidung von Quecksilber aus Rauchgasen von Hochtemperaturanlagen, insbesondere von Kraftwerken und Abfallverbrennungsanlagen, welches darin besteht, dass der, gegebenenfalls mehrstufigen, Feuerung und/oder dem Rauchgas in einem der Feuerung nachgeschalteten Anlagenteil Brom und/oder eine bromhaltige Verbindung und/oder eine Mischung verschiedener bromhaltiger Verbindungen zugeführt wird, wobei die Temperatur beim Kontakt der bromhaltigen Verbindung mit dem Rauchgas mindestens 500°C, vorzugsweise mindestens 800°C, beträgt, die Verbrennung in Gegenwart einer schwefelhaltigen Verbindung, insbesondere Schwefeldioxid, gegebenenfalls unter Zusatz von Schwefel und/oder einer schwefelhaltigen Verbindung und/oder einer Mischung verschiedener schwefelhaltiger Verbindungen erfolgt und anschließend das Rauchgas einer, gegebenenfalls mehrstufigen, Reinigung zur Abtrennung von Quecksilber aus dem Rauchgas unterzogen wird, welche eine Nasswäsche und/oder eine Trockenreinigung umfasst.

[0007] Die Abscheidung von Quecksilber aus Rauchgasen in einer der Verbrennung oder einem ähnlichen Hochtemperaturprozess nachgeschalteten Rauchgasreinigung hängt entscheidend davon ab, in welcher Speziesform das Quecksilber vor Eintritt in die Rauchgasreinigung vorliegt. Ein möglichst hoher Anteil an ionischem Quecksilber ist von Vorteil, da das ionische Quecksilber gut wasserlöslich, also auswaschbar, sowie an diversen Adsorbentien gut adsorbierbar ist. Der Zusatz von Brom bzw. bromhaltigen Verbindungen zur Feuerung bewirkt unter den gegebenen Bedingungen eines Hochtemperaturprozesses oder dgl. in Gegenwart einer schwefelhaltigen Verbindung, insbesondere in Gegenwart von Schwefeldioxid, eine weitgehende, im Wesent-

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lichen vollständige, Oxidation des Quecksilbers und erlaubt daher eine starke Abscheidung des Quecksilbers aus Rauchgasen.

[0008] Unter Hochtemperaturanlagen werden im Sinne der vorliegenden Erfindung insbesondere Abfallverbrennungsanlagen, beispielsweise Hausmüll-, Sondermüll- und Klärschlammverbrennungsanlagen, sowie Kraftwerke, beispielsweise Steinkohle- oder Braunkohlekraftwerke, aber auch sonstige Anlagen für Prozesse hoher Temperatur, wie z.B. das Zementbrennen, sowie Abfall mitverbrennende Hochtemperaturanlagen oder kombinierte (mehrstufige) Hochtemperaturanlagen, wie z.B. Kraftwerke oder Zementdrehrohröfen mit vorgeschalteter Abfallpyrolyse oder Abfallvergasung, verstanden. Die Dimension der Hochtemperaturanlage spielt für das erfindungsgemäße Verfahren keine Rolle. Vorteilhaft an dem erfindungsgemäßen Verfahren ist gerade, dass es auf unterschiedliche Typen von Hochtemperaturanlagen und auf Hochtemperaturanlagen von unterschiedlicher Größenordnung anwendbar ist. Dies schließt Anlagen mit einem Rauchgasvolumenstrom von nur $15 \cdot 10^3$ m^3 i. N. tr./h, beispielsweise zur Klärschlammverbrennung, oder von $50 \cdot 10^3$ m^3 i. N. tr./h, z.B. in Sondermüllverbrennungsanlagen, oder von $150 \cdot 10^3$ m^3 i. N. tr./h, z.B. in der Hausmüllverbrennung, ebenso ein wie Großkraftwerke mit z.B. $2–3 \cdot 10^6$ i. N. tr./h.

[0009] Für das erfindungsgemäße Verfahren ist es unerheblich, in welcher Form das zugeführte Brom vorliegt. Es ist möglich, freies oder organisch gebundenes oder anorganisch gebundenes Brom einzusetzen. Das Brom oder die bromhaltigen Verbindungen können einzeln oder im Gemisch zugeführt werden. Besonders bevorzugt wird eine wässrige Lösung von Bromwasserstoff oder ein Alkalibromid, insbesondere Natriumbromid, oder eine wässrige Lösung des Alkalibromids verwendet. Durch diese Ausführungsform ist das Verfahren wirtschaftlich besonders interessant, da die Kosten für zusätzliche Betriebsmittel niedrig gehalten werden können. Weiterhin bevorzugt ist eine Ausführungsform, in der die bromhaltige Verbindung bzw. das Gemisch verschiedener bromhaltiger Verbindungen aus bromreichen Abfallstoffen, wie z.B. niedrig- oder hochhalogenierte Flüssigabfälle, bestehen, welche Teil des Verbrennungsguts sind oder dem Verbrennungsgut, beispielsweise Sondermüll, beigemengt werden.

[0010] Das erfindungsgemäße Verfahren findet in Gegenwart einer schwefelhaltigen Verbindung statt. Die Zugabe einer bromhaltigen Verbindung nach dem erfindungsgemäßen Verfahren führt zu einer Gasphasenreaktion zwischen Quecksilber und Brom in Gegenwart von Schwefeldioxid. Da bei den im Sinne dieser Erfindung üblichen Verbrennungs- und sonstigen Hochtemperaturprozessen in der Regel Schwefeldioxid entsteht, ist für das erfindungsgemäße Verfahren in der Regel ein ausreichendes Angebot einer schwefelhaltigen Verbindung vorhanden. Ein ausreichendes Angebot im Sinne dieser Erfindung liegt vor, wenn unter Zugabe einer bromhaltigen Verbindung zu der Feuerung der Gehalt an Schwefeldioxid im Rauchgas vor der Rauchgasreinigung deutlich größer als Null ist. Wird bei einem Verbrennungsprozess jedoch kein oder nicht ausreichend Schwefeldioxid gebildet, muss dem Prozess eine schwefelhaltige Verbindung zugeführt werden. Dies kann in Form von freiem oder gebundenem Schwefel, beispielsweise Schwefelgranulat, Abfallschwefelsäure oder andere schwefelreiche Abfallstoffe, erfolgen. Darüber hinaus kann insbesondere auch zur Minderung eines zu hohen Gehalts an freien Halogenen im Rauchgas eine schwefelhaltige Verbindung zugesetzt werden, wenn z.B. mehr bromhaltige Verbindung zugeführt worden ist, als zur Oxidation des vorliegenden Quecksilbers notwendig ist. Die Zudosierung einer schwefelhaltigen Verbindung kann z.B. nach dem in der bisher unveröffentlichten Patentanmeldung DE 10131464 beschriebenen Verfahren zur korrosions- und emissionsarmen Mitverbrennung hochhalogenierter Abfälle in Abfallverbrennungsanlagen erfolgen. Nach diesem Verfahren wird in den primären und/oder sekundären Feuerraum Schwefel oder ein entsprechender Schwefelträger geregelt zudosiert. Die Regelung der Schwefelmenge erfolgt im Wesentlichen proportional zur mit den Abfällen eingetragenen aktuellen Halogengesamtfracht im Kesselrauchgas. Der zudosierte Schwefel verbrennt im Feuerraum zu Schwefeldioxid, was kesselintern zu einer weitgehenden Unterdrückung zwischenzeitlich gebildeter freier Halogene im Kesselrauchgas sowie nachfolgend zur stabilen Halogeneinbindung in der alkalischen Wäsche führt. Die Zudosierung von Schwefel wird so geregelt, dass der Soll-Schwefeldioxidgehalt im Rauchgas am Kesseleingang oder der Soll-Schwefeldioxidrestgehalt am Kesselende, d.h. im Kesselrohgas vor einer beispielsweise nassen Rauchgasreinigung, im stationären Betriebszustand über einen einfachen primären Regelkreis eingehalten werden.

[0011] Wird speziell Natriumbromid der Feuerung zugesetzt, so ist ein Mehrverbrauch an Schwefeldioxid zu beobachten, welcher auf die Sulfatierung des Natriumbromids im Hochtemperaturbereich zurückzuführen ist.

[0012] Andererseits stellt ein relativ hoher Gehalt einer schwefelhaltigen Verbindung, insbesondere an Schwefeldioxid, im Rauchgas keinen Nachteil für das erfindungsgemäße Verfahren dar. Ein hoher Gehalt an Schwefeldioxid kann beispielsweise bei der Verbrennung von Steinkohlen auftreten, welche üblicherweise zwischen 0,5 und 1 Gew.-% Schwefel enthalten, oder bei einer geregelten Zudosierung einer schwefelhaltigen Verbindung, welche zur Unterdrückung zwischenzeitlich gebildeter freier Halogene (s.o.) zugesetzt wird. Unter den gegebenen Bedingungen eines Hochtemperaturprozesses findet auch in Gegenwart von überschüssigem Schwefeldioxid die Oxidation des Quecksilbers statt, welche nach dem erfindungsgemäßen Verfahren durch den Zusatz einer oder mehrerer bromhaltiger Verbindungen erzielt wird. Darin liegt ein besonderer Vorteil des vorliegenden Verfahrens, denn die Oxidation des Quecksilbers durch den Zusatz von bromhaltigen Verbindun-

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gen zeigt sich – anders als durch den Zusatz von chlorhaltigen Verbindungen – weitgehend unempfindlich gegenüber einem Überschuss an Schwefeldioxid.

[0013] Die Zugabe einer bromhaltigen Verbindung sowie gegebenenfalls einer schwefelhaltigen Verbindung erfolgt erfindungsgemäß zur Feuerung und/oder zum Rauchgas in einem der Feuerung nachgeschalteten Anlagenteil, wobei die Temperatur beim Kontakt der bromhaltigen Verbindung mit dem Rauchgas mindestens 500°C, vorzugsweise mindestens 800°C, beträgt. Die bromhaltige Verbindung, z.B. Natriumbromid, kann entweder in fester, z.B. als Salz, oder flüssiger Form, z.B. als wässrige Lösung, dem zu verfeuernden Abfallgemisch, der Kohle oder dgl. vor der Feuerung beigemengt werden. Die Zugabe kann auch in einen der Feuerung vorgeschalteten Anlagenteil erfolgen, beispielsweise einer Pyrolysetrommel, welche z.B. dem thermischen Aufschluss mitverbrannter Abfallstoffe dient, oder einer Kohlemühle. Die Verbindung kann ebenso während des Verbrennungsprozesses zugeführt werden. Umfasst die Feuerung mehrere Stufen, z.B. eine primäre und eine sekundäre Feuerung, kann die bromhaltige Verbindung in eine oder beide Brennkammern, beispielsweise in den Drehrohrofen und/oder die Nachbrennkammer, ebenfalls in fester oder flüssiger Form eingeführt werden. Vorzugsweise wird eine wässrige Lösung der Verbindung in einer der Brennkammern eingedüst. Ferner kann die Zugabe auch nach der Verbrennung, z.B. in einem nachgeschalteten Abhitzekessel, erfolgen, sofern die Temperatur des Rauchgases ausreichend hoch ist, d.h. mindestens 500°C, insbesondere mindestens 800°C, beträgt. In anderen Hochtemperaturprozessen, wie z.B. dem Zementbrennen, wird beispielsweise der heiße Ofenkopf des Zementdrehrohrofens und/oder die befeuerte Entsäuerungsstufe des nachgeschalteten Zementrohmehlvorwärmers mit der bromhaltigen Verbindung bestückt.

[0014] In einer weiteren Ausführungsform des erfindungsgemäßen Verfahrens ist es auch möglich, die bromhaltige Verbindung, beispielsweise eine wässrige Lösung von Bromwasserstoff oder von Natriumbromid, fein dispergiert der Verbrennungsluft und/oder gegebenenfalls einem rückgeführten Teilstrom, insbesondere rückgeführtem Rauchgas, rückgeführter Asche und rückgeführten Flugstäuben, zuzuführen.

[0015] Um eine möglichst vollständige, insbesondere 100%ige, Oxidation des Quecksilbers durch die Zugabe einer bromhaltigen Verbindung zu erzielen, wird die bromhaltige Verbindung bevorzugt in einem Massenverhältnis von Brom. zu Quecksilber im Bereich von 10^2 bis 10^4 zugegeben. Wird die bromhaltige Verbindung in großem Überschuss zugefügt, wirkt sich dies nicht nachteilig auf das erfindungsgemäße Verfahren aus. Ein zu großer Überschuss muss jedoch vermieden werden, nicht zuletzt aus Kostengründen. Gegebenenfalls müssen zwischenzeitlich gebildete freie Halogene, wie z.B. freies Brom, durch die Zugabe einer schwefelhaltigen Verbindung unterdrückt bzw. stabil eingebunden werden (s.o.), da auch Bromemissionen in der Regel gesetzlich festgelegten Grenzwerten unterliegen.

[0016] Die Oxidation von Quecksilber kann prinzipiell auch durch chlorhaltige oder jodhaltige Verbindungen erfolgen. Es zeigt sich jedoch, dass bromhaltige Verbindungen Quecksilber unter den gegebenen Bedingungen von Hochtemperaturprozessen, wie der Temperatur, und insbesondere auch bei einem hohen Gehalt an Schwefeldioxid (s.o.) wirksamer oxidieren als chlorhaltige Verbindungen. Jodhaltige Verbindungen oxidieren im Vergleich zu bromhaltigen Verbindungen Quecksilber wirksamer. Aus wirtschaftlichen Gesichtspunkten werden in dem erfindungsgemäßen Verfahren allerdings bromhaltige Verbindungen bevorzugt eingesetzt. Gegebenenfalls in den Abfallstoffen, z.B. in Sondermüll, vorliegende chlor- oder jodhaltige Verbindungen tragen demnach zur Oxidation des Quecksilbers bei. In einer bevorzugten Ausführungsform erfolgt das erfindungsgemäße Verfahren zusätzlich zu den bromhaltigen Verbindungen in Gegenwart von Chlor und/oder Jod und/oder einer chlorhaltigen Verbindung und/oder einer jodhaltigen Verbindung und/oder einem Gemisch solcher Verbindungen. Die chlorhaltige und/oder jodhaltige Verbindung kann beispielsweise in Form von chlor- oder jodreichen Abfallstoffen in Ergänzung zu oder unter teilweisem Ersatz der zugesetzten bromhaltigen Verbindung zugeführt werden.

[0017] Nach dem erfindungsgemäßen Verfahren folgt nach der Verbrennung oder einem ähnlichen Hochtemperaturprozess unter Zusatz einer bromhaltigen Verbindung eine Reinigung des Rauchgases, wodurch das oxidierte Quecksilber möglichst weitgehend aus dem Rauchgas abgeschieden wird. Verschiedene Rauchgasreinigungsverfahren zur Abscheidung von u.a. ionischem Quecksilber sind aus dem Stand der Technik bekannt. Sie basieren entweder auf einer Nasswäsche oder einer Trockenreinigung oder einer Kombination beider und kann ggf. mehrstufig sein. Die Nasswäsche umfasst z.B. eine saure Wäsche, welche beispielsweise in einer mit Kreislaufwaschwasser bedüsten Quenche, einem Druckdüsen- oder Rotationszerstäuberwäscher oder einem Füllkörperwäscher erfolgt. Eine Wäsche kann aber ggf. auch nur schwachsauer oder alkalisch ausgeführt sein, beispielsweise im Fall niedriger Chlorwasserstofffrachten, aber hoher Schwefeldioxidfrachten.

[0018] In einer bevorzugten Ausführungsform umfasst die Rauchgasreinigung eine mehrstufige nasse Rauchgaswäsche mit mindestens einer stark sauren (pH kleiner 1) und/oder mindestens einer schwach sauren (pH 3–4) und/oder mindestens einer alkalischen Waschstufe.

[0019] Die Rauchgasreinigung kann auch eine auf der Adsorption von ionischen Quecksilberverbindungen basierende Trockenreinigung umfassen. Eine solche Reinigung kann beispielsweise durch quasitrockene Entschwefelung in einem Sprühtrockner durchgeführt werden, welcher mit einer Kalkmilch-Kohle-Suspension beaufschlagt ist, oder mit Hilfe von Festbettadsorbern, z.B. auf Basis von körniger Aktivkohle oder von Herdofen-

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koks oder von Mischungen derartiger Adsorbentien mit gekörntem Kalk, oder mit Hilfe von Flugstromadsorbern, z.B. elektrostatischen Gasreinigungsfiltern (EGR-Filter), oder mit Hilfe von Gewebefiltern, welche mit einem eingeblasenen feinpulvrigen Kalkhydrat-Aktivkohle- oder Kalkhydrat-Herdofenkoks-Gemisch beaufschlagt sind. Auch Zeolithe eignen sich zur Abscheidung von quecksilberhaltigen Verbindungen. Im Bezug auf die trockene Rauchgasreinigung zeigt sich ein weiterer Vorteil des erfindungsgemäßen Verfahrens. Die Anwendung des Verfahrens ist insbesondere auch für jene Hochtemperaturanlagen interessant, welche über keine nasse Rauchgasreinigung, sondern ausschließlich über eine trockene Reinigung mit einer Quecksilbersorptionsstufe verfügen. Quecksilbercromid $HgBr_2$ adsorbiert stärker an Trockensorbentien als Quecksilberchlorid $HgCl_2$. Beispielsweise verstärkt sich die Adsorption von Quecksilber am Flugstaub von EGR-Filtern.

[0020] In einer bevorzugten Ausführungsform umfasst die Rauchgasreinigung daher mindestens eine trockene oder quasitrockene, auf Adsorption basierende Reinigungsstufe, insbesondere unter Verwendung von elektrostatischen oder filternden Staubabscheidern.

[0021] Des weiteren werden die mit Quecksilber beladenen Flugstäube aus gegebenenfalls vorhandenen Staubabscheidern zwecks Quecksilberentfrachtung vorzugsweise thermisch nachbehandelt, insbesondere in einer auf Temperaturen von mindestens 200°C beheizten Drehtrommel.

[0022] Bevorzugt wird in dem erfindungsgemäßen Verfahren der Quecksilbergehalt des Rauchgases, insbesondere der Gehalt an metallischem Quecksilber, nach der Rauchgasreinigung kontinuierlich gemessen und anhand des gemessenen Quecksilbergehalts die Menge an zugeführtem Brom und/oder bromhaltigen Verbindungen und/oder der Mischung bromhaltiger Verbindungen sowie gegebenenfalls Schwefel und/oder schwefelhaltigen Substanzen und/oder der Mischung schwefelhaltiger Substanzen geregelt. Ein relativ hoher Gehalt an metallischem Quecksilber im Rauchgas ist ein Indikator dafür, dass die Oxidation des Quecksilbers unvollständig verläuft und damit das Quecksilber in der Rauchgasreinigung unvollständig abgetrennt wird. Um Quecksilber möglichst vollständig zu oxidieren, muss in einem solchen Fall mehr bromhaltige Verbindung zugeführt werden. Zusätzlich kann der Gehalt an ionischem Quecksilber nach der Rauchgasreinigung gemessen werden und daraus der Abscheidegrad des ionischen Quecksilbers in der Rauchgasreinigung bestimmt werden. Der Gehalt an metallischem Quecksilber und ggf. an Quecksilber insgesamt im Kesselrohgas kann nach entsprechender Gasaufbereitung beispielsweise mit Hilfe eines Differenz-Absorptionsphotometers gemessen werden. Die kontinuierliche Messung von metallischem Quecksilber und ggf. auch von Quecksilber insgesamt im Reingas nach der nassen und/oder trockenen Rauchgasreinigung erfolgt vorzugsweise vor einer gegebenenfalls vorhandenen, nachgeschalteten SCR-Entstickungsanlage (SCR: selective catalytic reduction), da der metalloxidreiche Festbettkatalysator erhebliche Mengen an metallischem Quecksilber adsorbiert.

[0023] Nachfolgend wird die Erfindung anhand der Beispiele unter Bezugnahme auf die anliegenden Figuren näher erläutert. Es zeigen:

[0024] **Fig.** 1 ein Schema einer Sondermüllverbrennungsanlage

[0025] **Fig.** 2 ein Diagramm, welches den Gehalt von metallischem Quecksilber (Hg_{met}) im gewaschenen Kesselrauchgas, d.h. im Reingas, nach der Nasswäsche in µg/m³ i. N. tr. (Kurve 21, linke Ordinate) sowie den Gesamtgehalt von Brom (Br_{ges}) im Kesselrauchgas in mg/m³ i. N. tr. (Kurve **22**, rechte Ordinate) in Abhängigkeit von der Zeit darstellt,

[0026] **Fig.** 3 ein Diagramm, welches den Gesamtgehalt von Quecksilber (Hg_{ges}) im Kesselrauchgas, d.h. zugleich im Kesselrohgas, vor der Nasswäsche in µg/m³ i. N. tr. (Kurve 31, linke Ordinate) sowie den Gehalt von metallischem Quecksilber (Hg_{met}) im Reingas nach der Nasswäsche in µg/m3 i. N. tr. (Kurve 32, rechte Ordinate) in Abhängigkeit von der Zeit darstellt,

[0027] **Fig.** 4 ein Diagramm, welches den Gesamtgehalt von Brom (Br_{ges}) im Kesselrauchgas, d.h. zugleich im Kesselrohgas, vor der Nasswäsche in mg/m³ i. N. tr. (Kurve 41, linke Ordinate) sowie den Gehalt von metallischem Quecksilber (Hg_{met}) im Reingas nach der Nasswäsche in µg/m³ i. N. tr. (Kurve 42, rechte Ordinate) als Funktion der Zeit darstellt,

[0028] **Fig.** 5 ein Diagramm, welches das Massenverhältnis von Brom zu Quecksilber im Kesselrauchgas (Kurve 51, linke Ordinate) sowie den erzielten Gesamtabscheidegrad von Quecksilber in der mehrstufigen Nasswäsche in (Kurve 52, rechte Ordinate) als Funktion der Zeit darstellt,

[0029] **Fig.** 6 ein Diagramm, welches das Gewichtsverhältnis von metallischem Quecksilber zur Summe aus metallischem und ionischem Quecksilber (Hg_{met}/Hg_{ges}), d.h. den Hg_{met}-Speziesanteil im Kesselrohgas, in Gew.-% als Funktion des Gesamtgehaltes an Chlor (Kurve 61) bzw. an Brom (Kurve 62) im Kesselrohgas in mg/m³ i. N. tr. darstellt,

[0030] **Fig.** 7 ein Diagramm, welches den Gesamtgehalt an Quecksilber (Hg_{ges}) im entstaubten Rohgas nach der elektrostatischen Gasreinigung (Kurve 71, linke Ordinate) bzw. den Gehalt an metallischem Quecksilber (Hg_{met}) nach der elektrostatischen Gasreinigung (Kurve 72, linke Ordinate) sowie die durch Quecksilberzudosierung herbeigeführte Anhebung des Gesamtgehalts an Quecksilber (Hg_{ges}) im Kesselrauchgas (Kurve 73, rechte Ordinate) als Funktion der Zeit darstellt,

[0031] **Fig.** 8 ein Diagramm, welches das Gewichtsverhältnis von metallischem Quecksilber (Hg_{met}) zu der Summe aus metallischem und ionischem Quecksilber (Hg_{ges}), d.h. den Hg_{met}-Speziesanteil (Hg_{met}/Hg_{ges}), im

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entstaubten Kesselrohgas nach der elektrostatischen Gasreinigung in Gew.-% (Kurve 82) sowie den Gesamtgehalt an Brom (Br_{ges}) im Kesselrauchgas in mg/m^3 i. N. tr. (Kurve 81) als Funktion der Zeit darstellt.
[0032] **Fig.** 9 ein Schema eines Industriekraftwerks mit zwei Schmelzkammerkesseln.

Beispiele

[0033] Die Beispiele 1–4 wurden in einer dem Schema in **Fig.** 1 entsprechenden Sondermüllverbrennungsanlage der Bayer AG in Leverkusen durchgeführt. Der Drehrohrofen **3** als primärer Feuerraum wird mit Feststoffabfällen aus dem Bunker **1** über einen Krangreifer **2**, mit Flüssigabfällen aus einem Flüssigabfalltank sowie mit Abfallgebinden über eine Gebindeaufgabe befeuert. Auch die Nachbrennkammer **4** als sekundärer Feuerraum wird mit Flüssigabfällen befeuert. Das Rauchgas wird über den Abhitzekessel **5** abgekühlt und anschließend, als sogenanntes Kesselrohgas, der nassen Rauchgasreinigung (mehrstufige Wasche) zugeführt, welche eine Quenche **6**, einen sauren Rotationszerstäuberwäscher **7**, einen alkalischen Rotationszerstäuberwäscher **8** und eine elektrostatische Gasreinigung mit Wasserdampf-Teilkondensation **9** (KEGR) umfasst. Über Saugzüge **10** gelangt das gewaschene Rauchgas, als sogenanntes Reingas, in die nachgeschaltete katalytische Entstickungsanlage **11** (selektive katalytische Entstickung des Reingases mittels Ammoniak) und wird von dort über den Kamin **12** emittiert. Der Gehalt an metallischem Quecksilber (Hg_{met}) sowie gegebenenfalls der Gesamtgehalt an Quecksilber (Hg_{ges}) im gewaschenen Reingas nach KEGR wurde – nach entsprechender Aufbereitung – an der Messstelle **16** mit Hilfe eines Differenz-Absorptionsphotometers kontinuierlich bestimmt. Der Gehalt an Quecksilber insgesamt (Hgges) im emittierten Reingas wurde an der Messstelle **17**, d.h. in einer Kaminhöhe von 22 m, quasikontinuierlich bestimmt durch Amalgamierung an einer periodisch ausgeheizten Goldfolie mit nachfolgendem Differenz-Absorptionsphotometer.
[0034] Beispiel **5** beschreibt die Anwendung des erfindungsgemäßen Verfahrens in einem Kohlekraftwerk der Bayer AG in Uerdingen, welches im Wesentlichen einen Schmelzkammerkessel und eine kraftwerkstypische Rauchgasreinigung, bestehend aus einer trockenen elektrostatischen Gasreinigung (EGR), einem schwachsauren, auf Kalkstein basierten Nasswäscher zur Rauchgasentschwefelung sowie einer SCR-Entstickungsanlage (SCR: selective catalytic reduction), umfasst.

Ausführungsbeispiel

Beispiel 1

[0035] Über einen Zeitraum von 116 Minuten wurde dem sekundären Feuerraum (Nachbrennkammer **4**) über die Schauluke **15** eine Serie von Proben von metallischem Quecksilber in Kunststoffkapseln zugeführt (in Summe 3400 g, siehe Tabelle 1). Die Zugabe erfolgte in Abständen von ca. 5–10 Minuten mit zunehmender Quecksilbermenge. Das eingeworfene Quecksilber verdampft innerhalb von ca. 2–4 Minuten; demzufolge können die im Kesselrauchgas mit einem Volumenstrom von ca. 45·103 m^3 i. N. tr./h aufgetretenen momentanen Spitzenkonzentrationen an Quecksilber abgeschätzt werden. Die Abschätzung liefert zum Versuchsende hin Spitzenkonzentrationen von mehr als $130 \cdot 10^3$ µg/m^3 i. N. tr.

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Tabelle 1: Zugabe der Hg-Proben

Uhrzeit	Hg-Menge [g]	Uhrzeit	Hg-Menge [g]
9:24	5	10:32	180
9:32	10	10:37	200
9:38	15	10:43	220
9:49	20	10:48	240
9:54	40	10:53	260
9:59	60	10:58	280
10:04	80	11:03	300
10:09	100	11:08	310
10:15	120	11:13	320
10:20	140	11:20	340
10:26	160		
Versuchsdauer [min]		**Hg-Gesamtmenge [g]**	
116		**3400**	

[0036] Während des Versuchszeitraums wurde durch Mitverbrennung eines hochbromierten Flüssigabfalls (Zugabe zum Drehrohrofenkopf) im Kesselrauchgas von $45 \cdot 10^3$ m^3 i.N. tr./h ein Bromgehalt von ca. $4 \cdot 10^3$ mg/m^3 i. N. tr. aufrechterhalten, wie die Kurve 22 (rechte Ordinate) in **Fig.** 2 zeigt (Bestimmung anhand des Durchsatzes und Bromgehaltes des hochbromierten Flüssigabfalls). Der Rest-SO_2-Gehalt im Kesselrohgas vor der Quenche war dabei mittels Zudosierung von Schwefelgranulat zum Drehrohrofenkopf auf $5{,}5 \cdot 10^3$ mg/Nm^3 i. N. tr. ungewöhnlich hoch eingestellt (direkte SO_2-Messung im Kesselrohgas vor der Quenche). Damit war sichergestellt, dass für das erfindungsgemäße Verfahren ein ausreichendes Angebot an Schwefeldioxid vorhanden war. Das sonstige Verbrennungsgut bestand aus Festabfällen und schwachchlorierten Lösemitteln. Vor, während und nach der Quecksilberzugabe wurde an der Messstelle **16**, d.h. nach der Rauchgasreinigung, der Gehalt an Quecksilber im Rauchgas gemessen. Wie die Kurve **21** (linke Ordinate) in **Fig.** 2 zeigt, steigt trotz der Zugabe von erheblichen Quecksilbermengen der Gehalt an metallischem Quecksilber, welches durch die Wäsche durchschlägt, praktisch nicht an.

[0037] Darüber hinaus sind in Tabelle 2 die momentanen Austragungsströme von Quecksilber um 11:30 Uhr, d.h. kurz nach der Zugabe der letzten Quecksilberprobe und damit zum Zeitpunkt der höchsten Quecksilberkonzentration, aufgeführt, welche mit den Wäscherabwässern der nassen Rauchgasreinigung ausgetragen wurden. Umfangreiche abwasserseitige Messungen belegen, dass ca. 99,93 % des insgesamt ausgetragenen Quecksilbers als ionisches Quecksilber mit dem Abwasser der stark sauren Quenche (pH-Wert kleiner 1) und ca. 0,066 % mit dem Abwasser des alkalischen Rotationszerstäuberwäschers (pH-Wert ca. 7,5) ausgetragen wurden. Der geringe, nicht ausgewaschene Rest von nur 0,004 % des insgesamt ausgetragenen Quecksilbers wurde mit dem gewaschenen Reingas als metallisches Quecksilber ausgetragen. Nachweislich war im gewaschenen Reingas praktisch kein Hg_{ion} zu finden (Hg_{ion} = Null, d.h. vollständige Auswaschung des ionischen Quecksilbers und somit $Hg_{ges} = Hg_{met}$).

Tabelle 2: Momentane Quecksilber-Austragungsströme [g/h] um 11:30 Uhr

Quenche (einschließlich dem sauren Rotationszerstäuberwäscher)	1931
(Saurer Rotationszerstäuberwäscher, dessen Abwasser zur Quenche rückgeführt wird)	(468)
Alkalischer Rotationszerstäuberwäscher	1,32
Gewaschenes Reingas nach KEGR	0,069

Beispiel 2:

[0038] Über einen Zeitraum von 130 Minuten wurde dem sekundären Feuerraum (Nachbrennkammer **4**) über eine Düse in der Nachbrennkammerdecke kontinuierlich eine wässrige $HgCl_2$-Lösung zugeführt. Dabei wurde die zudosierte Menge in Zeitschritten von etwa 5 Minuten angehoben. **Fig.** 3 zeigt den so herbeigeführten Anstieg der Konzentration von Quecksilber im Kesselrauchgas in der Zeit zwischen ca. 10:45 und 13:00 Uhr. Das eindosierte Quecksilber wird in der Nachbrennkammer unverzüglich als metallisches Quecksilber Hg_{met} freigesetzt. Die Gesamtkonzentration von Quecksilber im Kesselrauchgas stieg auf diese Weise bis auf Werte von $18 \cdot 10^3$ µg/m³ i. N. tr. an (Kurve **31** und linke Ordinate). Die Hg-Konzentration im Kesselrauchgas wurde aus dem zugegebenen Quecksilbermengenstrom und dem betrieblich gemessenen Rauchgasvolumenstrom berechnet. Während des Versuchszeitraums wurde durch Mitverbrennung eines hochbromierten Flüssigabfalls (Zugabe über einen Brenner am Drehrohrofenkopf) im Kesselrauchgas von $45 \cdot 10^3$ m³ i.N. tr./h ein Bromgehalt von ca. $9 \cdot 10^3$ mg/m³ i. N. tr. aufrechterhalten (Bestimmung anhand des Durchsatzes und Bromgehalt des mitverbrannten hochbromierten Flüssigabfalls). Der Rest-SO_2-Gehalt im Kesselrohgas vor Quenche war dabei mittels Zudosierung von Schwefelgranulat zum Drehrohrofenkopf auf ca. $4 \cdot 10^3$ mg/Nm³ i. N. tr. eingestellt (direkte SO_2-Messung im Kesselrohgas vor der Quenche).

[0039] Im Zeitraum zwischen ca. 11:00 und 13:00 Uhr wurde im gewaschenen Reingas nach der KEGR eine Konzentration von metallischem Quecksilber von weniger als 10 µg/m³ i. N. tr. beobachtet. Nachweislich war im gewaschenen Reingas auch hier praktisch kein Hg_{ion} zu finden (Hg_{ion} = Null, d.h. vollständige Auswaschung des ionischen Quecksilbers und somit $Hg_{ges} = Hg_{met}$). Bei einem kurzzeitigen Ausfall der Bromzugabe gegen 13:05 Uhr stieg die Konzentration von Hg_{met} sprunghaft auf ca. 800 µg/m³ i. N. tr. an und kehrte aber unverzüglich auf ihren niedrigen Ausgangswert von weniger als 10 µg/m³ i. N. tr. zurück, als die Bromzugabe wieder einsetzte (Kurve **32** und rechte Ordinate).

Beispiel 3:

[0040] In der Zeit zwischen ca. 8:30 und 14:45 Uhr, d.h. über einen Zeitraum von 675 Minuten, wurde dem sekundären Feuerraum (Nachbrennkammer **4**) über eine Düse in der Nachbrennkammerdecke kontinuierlich eine wässrige $HgCl_2$-Lösung zugeführt. Dabei wurde nunmehr aber der zudosierte Hg-Mengenstrom konstant gehalten, entsprechend einer Konzentration von Quecksilber im Kesselrauchgas von ca. $9{,}6 \cdot 10^3$ µg/m³ i. N. tr.

[0041] In diesem Versuchszeitraum (vgl. **Fig.** 4 bzw. **Fig.** 5) wurde Brom in Form eines hochbromierten Flüssigabfalls über einen Brenner am Drehrohrofenkopf zugegeben, jedoch wurde die zugegebene Brommenge schrittweise verringert, womit der Bromgehalt im Kesselrauchgas stufenweise von ca. $9 \cdot 10^3$ auf ca. $3 \cdot 10^3$ mg/m³ i. N. tr. abgesenkt wurde (Kurve **41** in **Fig.** 4 und linke Ordinate). Der mittels Zudosierung von Schwefelgranulat herbeigeführte Rest-SO_2-Gehalt im Kesselrohgas war mit ca. $4{,}3 \cdot 10^3$ mg/m³ i. N. tr in diesem Versuchszeitraum wiederum sehr hoch gewählt worden. Neben dem hochbromierten Flüssigabfall wurde auch ein chlorierter Flüssigabfall mitverbrannt.

[0042] Wie **Fig.** 4 bzw. **Fig.** 5 zu entnehmen ist, war der Gehalt an metallischem Quecksilber im gewaschenen Reingas nach KEGR deutlich kleiner als 2 µg/m³ i. N.

[0043] tr. (Kurve **42** in **Fig.** 4 und rechte Ordinate). Nachweislich war im gewaschenen Reingas auch hier praktisch kein Hg_{ion} zu finden (Hg_{ion} = Null, d.h. vollständige Auswaschung des ionischen Quecksilbers und somit $Hg_{ges} = Hg_{met}$). Entsprechend war der Abscheidegrad von Quecksilber in der Nasswäsche deutlich größer als 99,98 % (Kurve **52** in **Fig.** 5 und rechte Ordinate), solange der Bromgehalt gößer als $3 \cdot 10^3$ mg/m³ i. N. tr.

(Kurve **41** und linke Ordinate) bzw. das Brom-Quecksilber-Massenverhältnis größer als 500 µg Brom/µg Quecksilber betrug (Kurve **51** in **Fig.** 5 und linke Ordinate). Gegen 13:30 Uhr sinkt der Bromgehalt im Rauchgas auf $3 \cdot 10^3$ mg/m^3 i. N. tr. bzw. das Brom-Quecksilber-Massenverhältnis auf ca. 335 µg Brom/µg Quecksilber. Dabei steigt die Konzentration von metallischem Quecksilber nach der Nasswäsche auf bis zu 20 µg/m^3 i. N. tr. (Kurve **42** in **Fig.** 4 und linke Ordinate) bzw. nimmt der Hg-Abscheidegrad auf 99,8 % ab (Kurve **52** in **Fig.** 5 und rechte Ordinate). Darüber hinaus führt eine kurzzeitige Unterbrechung der Chlor-Zugabe kurz nach 14:30 Uhr zu einer Spitzenkonzentration von metallischem Quecksilber nach der Wäsche von ca. 117 µg/m^3 i. N. tr. (Kurve **42** in **Fig.** 4 und linke Ordinate) bzw. zu einem kurzzeitigen Abfall des Abscheidegrades auf ca. 98,4 % (Kurve **51** in **Fig.** 5 und rechte Ordinate). Hieran wird die gegenüber Brom vergleichsweise geringe Wirkung von Chlor deutlich.

Beispiel 4:

[0044] **Fig.** 6 illustriert einen Versuch zum Vergleich der Wirkung von Brom und Chlor auf die Oxidation von Quecksilber im Kesselrauchgas der oben beschriebenen Sondermüllverbrennungsanlage. Dabei lagen ein durch Zudosierung von $HgCl_2$ eingestellter Hgges Gehalt von 130 µg/m^3 i. N. tr. bei einem durch Mitverbrennung chlorarmer Lösemittel eingestellten Chlorgehalt (Cl_{ges}) im Kesselrauchgas von $1{,}35 \cdot 10^3$ mg/m^3 i. N. tr. sowie einem durch Zudosierung von Schwefelgranulat eingestellten Restgehalt an Schwefeldioxid im Kesselrohgas von $1{,}5 \cdot 10^3$ mg/m^3 i. N. tr. vor. Der Messpunkt **63** zeigt den zunächst ohne Bromzusatz, d.h. allein durch Chlor, erzielten Hg_{met}-Speziesanteil von ca. 63 Gew.-% im Kesselrohgas vor der Nasswäsche. Die anlagenspezifische Kurve **61**, welche auf ca. 20 Betriebsversuchen an einer Sondermüllverbrennungsanlage mit Verbrennung hochchlorierter Flüssigabfälle basiert, zeigt, wie der Hgmet-Speziesanteil (Hg_{met}/Hg_{ges}) mit wachsendem Chlorgehalt Cl_{ges} im Kesselrauchgas zurückgeht.

[0045] Ausgehend von einem Hgmet-Speziesanteil von ca. 63 Gew.-% im Kesselrohgas vor der Nasswäsche (Messpunkt **63** mit Cl_{ges}-Gehalt als Abszisse bzw. Messpunkt **63'** mit Brges Gehalt als Abszisse) wurde dann in drei Schritten eine zunehmende Menge einer bromhaltigen Verbindung zudosiert (vgl. Pfeil **64**, der den Übergang von der Darstellung des Hg_{met}-Speziesanteil in Abhängigkeit von dem Cl_{ges} Gehalt zu der Darstellung in Abhängigkeit von dem Br_{ges}-Gehalt markiert). Dabei wurde der Bromgehalt im Kesselrauchgas von zunächst 0 mg/m^3 i. N. tr. (Messpunkt **63'** mit Br_{ges}-Gehalt als Abszisse) durch Zudosierung von wässriger Bromwasserstofflösung bzw. von wässriger Natriumbromidlösung (Eindüsung an der Nachbrennkammerdecke **14**, **Fig.** 1) in drei Schritten auf 50, 100 und 120 mg/m^3 i. N. tr. (Messpunkte **62** mit Brges Gehalt als Abszisse) angehoben. Dabei ging der Hg_{met}-Speziesanteil (Hg_{met}/Hg_{ges}) im Kesselrohgas vor der Nasswäsche – ausgehend von ca. 63 Gew.-% – bis auf 30 Gew.-% zurück.

[0046] Der Vergleich weist die deutlich effektivere Oxidation des Quecksilbers durch bromhaltige Verbindungen im Vergleich zu chlorhaltigen Verbindungen am Beispiel einer Sondermüllverbrennungsanlage nach. Um allein mittels Chlor einen Hg_{met}-Speziesanteil von nur 30 Gew.-% zu erreichen, müsste man den Cl_{ges}-Gehalt gemäß der Chlorierungskurve **61** auf $4 \cdot 10^3$ mg/m^3 i. N. tr. erhöhen. Statt dessen erreicht man dies mit nur 120 mg/m^3 i. N. tr. an Brom. Brom erscheint daher etwa 25-fach wirksamer als Chlor. Die Hg-Bromierungskurve **65** (Br_{ges}-Gehalt als Abszisse) entspricht unter Berücksichtigung dieses Faktors der vollständig vermessenen Hg-Chlorierungskurve **61** (Cl_{ges}-Gehalt als Abszisse). Gleiches gilt für im Fall von Kraftwerksrauchgasen, wo allerdings die anlagenspezifische Hg-Chlorierungskurve und die entsprechende Hg-Bromierungskurve **65** zu wesentlich niedrigeren Halogengehalten hin verschoben sind.

Beispiel 5:

[0047] **Fig.** 7 und **Fig.** 8 illustrieren Versuche zum Nachweis der Wirkung von Brom auf die Quecksilberabscheidung in einem Kohlekraftwerk der Bayer AG in Uerdingen (vgl. **Fig.** 9). In dem Kohlekraftwerk erfolgte ein Versuch mit Zudosierung von wässriger $HgCl_2$-Lösung und von wässriger NaBr-Lösung in den Feuerraum zum Nachweis der Wirkung von Brom auf die Hg-Oxidation. Das Kraftwerk umfasst zwei parallele Schmelzkammerkessel **91**, **91'** mit Temperaturen im Feuerraum um 1450°C. Die Schmelzkammerkesssel **91**, **91'** werden mit Kohle **92**, **92'** bestückt. Über die jeweiligen Lufvorwärmer **93**, **93'** wird den Schmelzlkammerkesseln **91**, **91'** Frischluft **94**, **94'** zugeführt. Das Kesselrohgas **95**, **95'** wird über elektrostatische Gasreiniger (EGR) **96**, **96'** dem gemeinsamen schwachsauren (pH = 5,3) Nasswäscher als Rauchgasentschwefelungsanlage (REA-Wäscher) **97** zugeführt. Das gewaschene Kesselrauchgas (Reingas) wird anschließend in zwei parallele katalytisch wirkende Entstickungsanlagen (SCR-Entstickungsanlage) **98**, **98'** überführt, bevor es über Kamine **100**, **100'** emittiert wird. Der in den EGR abgeschiedene Flugstaub **99**, **99'** wird zu 100 % in die Feuerung des jeweiligen Schmelzkammerkessels zurückgeführt. An der Messstelle **101** nach der EGR **96** wurden die Gehalte an Hg_{met} und Hgges im entstaubten Kesselrohgas kontinuierlich erfasst.

[0048] Es wurde kein Schwefel zudosiert. Der Gehalt an Schwefeldioxid im Kesselrauchgas von $1{,}3 \cdot 10^3$ i. N. tr. resultierte ausschließlich aus dem verbrannten Kohleschwefel selbst. Der Gesamtquecksilbergehalt im ent-

staubten Rohgas nach der EGR, d.h. vor der Nasswäsche, betrug zu Beginn bei reiner Kohleverbrennung (polnische Steinkohle) im Mittel nur 22,5 µg/m^3 i. N. tr., vgl. **Fig.** 7, Kurve **71** (Gesamtgehalt an Quecksilber Hg_{ges}) um 8:30 Uhr, der Gehalt an metallischem Quecksilber im Mittel nur 8,8 µg/m^3 i. N. tr., vgl. **Fig.** 7, Kurve **72** (Gehalt an metallischem Quecksilber Hg_{met}) um 8:30 Uhr. Die Zahnung beider Kurven **71**, **72** im 10-Minuten-Takt beruht auf der regelmäßigen Abklopfung der EGR; wodurch unmittelbar nach Abreinigung der Staubschichten im entstaubten Kesselrohgas hinter der EGR höhere Gehalte auftreten. Um 9:15 Uhr wurde mit der Zudosierung von Quecksilber (als wässrige $HgCl_2$-Löung) und um 10:30 dann auch mit der Zudosierung von Brom (als wässrige NaBr-Lösung) in den Feuerraum begonnen. Die Kurve **73** (**Fig.** 7, rechte Ordinate) stellt die durch Zudosierung von Quecksilber herbeigeführte Erhöhung des Hg_{ges}-Gehalts im Kesselrauchgas dar. Zwischen ca. 9:30 und 13:00 Uhr betrug die mittels $HgCl_2$-Zudosierung herbeigeführte Anhebung des Gesamtgehalts an Quecksilber im Rauchgas vor der EGR mindestens ca. 220 µg/m^3 i. N. tr. (Kurve **73**, rechte Ordinate). Die Kurve **81** in **Fig.** 8 stellt die durch Zudosierung von wässriger NaBr-Lösung herbeigeführte Erhöhung des Br-Gehalts im Kesselrauchgas dar. Um 10:30 Uhr wurde der Bromgehalt im Rauchgas vor der EGR zunächst um mindestens 75 mg/m^3 i. N. tr. angehoben und schrittweise wieder abgesenkt. Um 16:10 Uhr erfolgte eine neuerliche Anhebung des Bromgehalts um ca. 43 mg/m^3 i. N. tr. Wegen der Rückführung der Flugasche in die Schmelzkammerfeuerung und somit auch der Rückführung von an der Flugasche sorbiertem Quecksilber bzw. Brom handelt es sich hierbei um Mindestanhebungen, wie sie sich aus den zudosierten Mengenströmen und dem Rauchgasvolumenstrom (ca. $110 \cdot 10^3$ m^3 i. N. tr./h) ergeben. Die tatsächlichen Hg- und Br-Gehalte im Rohgas vor der EGR liegen demzufolge (Kreislauf zwischen Schmelzkammerfeuerung und EGR) etwas höher.

[0049] Die Kurven **71** bzw. **72** (linke Ordinate) in **Fig.** 7 zeigen, wie mit dem Zusatz der bromhaltigen Verbindung der Gehalt an Quecksilber im Rauchgas deutlich zurückgeht. Dies gilt einerseits für das ionische Quecksilber (Differenz zwischen Hg_{ges} und Hg_{met}), welches in Anwesenheit der bromhaltigen Verbindung vermehrt gebildet und offensichtlich am rückgeführten Flugstaub adsorbiert wird, und gilt andererseits aber stärker noch für das metallische Quecksilber, dessen Gehalt im entstaubten Rohgas nach EGR trotz des Zusatzes von Quecksilber annähernd auf den Ausgangsgehalt vor der Quecksilberzugabe zurückkehrt. Von 10:30 Uhr bis 13:00 Uhr (Ende der Br-Zudosierung) und weit darüber hinaus betrug der Gehalt an Hg_{met} weniger als 10 µg/m^3 i. N. tr. Erst mit dem Ende der neuerlichen Zugabe der Natriumbromidlösung gegen 19:00 Uhr steigt der Gehalt von Hg_{ges} deutlich an. Weiterhin zeigt die Kurve **82** in **Fig.** 8 den zunächst schlagartigen Rückgang des Speziesanteils an metallischem Quecksilber mit der Zugabe von Brom (Absenkung von ca. 40 Gew.-% auf ca. 10 Gew.-% gegen 10:30 Uhr). Entsprechendes zeigt sich nach ca. 17:00 Uhr bei der neuerlichen Quecksilber- und Bromzugabe an dem allmählichen Rückgang des Hg_{met}-Speziesanteils bis auf nur ca. 5 Gew.-% gegen 20:45 Uhr. Infolge der Hg-Zudosierung und der verstärkten Hg-Adsorption stieg der Hg-Gehalt im zur Schmelzkammer rückgeführten EGR-Flugstaub von anfänglich ca. 2–5 mg/kg im Laufe des Versuchs auf 55 mg/kg an.

Patentansprüche

1. Verfahren zur Abscheidung von Quecksilber aus Rauchgasen von Hochtemperaturanlagen, insbesondere von Kraftwerken und Abfallverbrennungsanlagen, **dadurch gekennzeichnet**, dass
– der, gegebenenfalls mehrstufigen, Feuerung und/oder dem Rauchgas in einem der Feuerung nachgeschalteten Anlagenteil Brom und/oder eine bromhaltige Verbindung und/oder eine Mischung verschiedener bromhaltiger Verbindungen zugeführt wird, wobei die Temperatur beim Kontakt der bromhaltigen Verbindung mit dem Rauchgas mindestens 500°C, vorzugsweise mindestens 800°C, beträgt,
– die Verbrennung in Gegenwart einer schwefelhaltigen Verbindung, insbesondere Schwefeldioxid, gegebenenfalls unter Zusatz von Schwefel und/oder einer schwefelhaltigen Verbindung und/oder einer Mischung verschiedener schwefelhaltiger Verbindungen erfolgt
– und anschließend das Rauchgas einer, gegebenenfalls mehrstufigen, Reinigung zur Abtrennung von Quecksilber aus dem Rauchgas unterzogen wird, welche eine Nasswäsche und/oder eine Trockenreinigung umfasst.

2. Verfahren nach Anspruch 1, dadurch gekennzeichnet, dass die bromhaltige Verbindung eine wässrige Lösung von Bromwasserstoff und/oder ein Alkalibromid, insbesondere Natriumbromid, und/oder eine wässrige Lösung von Alkalibromid ist.

3. Verfahren nach einem der Ansprüche 1 bis 2, dadurch gekennzeichnet, dass die bromhaltige Verbindung und/oder die Mischung bromhaltiger Verbindungen flüssige und/oder feste bromreiche Abfallstoffe sind.

4. Verfahren nach einem der Ansprüche 1 bis 3, dadurch gekennzeichnet, dass das Brom und/oder die bromhaltige Verbindung und/oder die Mischung bromhaltiger Verbindungen der Verbrennungsluft und/oder gegebenenfalls einem rückgeführten Teilstrom, insbesondere dem rückgeführtem Rauchgas und den rückgeführten Flugstäuben, zugesetzt wird.

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5. Verfahren nach einem der Ansprüche 1 bis 4, dadurch gekennzeichnet, dass das Massenverhältnis von Brom zu Quecksilber im Bereich von 10^2 bis 10^4 liegt.

6. Verfahren nach einem der Ansprüche 1 bis 5, dadurch gekennzeichnet, dass die Verbrennung zusätzlich in Gegenwart von Chlor und/oder einer chlorhaltigen Verbindung und/oder einer Mischung verschiedener chlorhaltiger Verbindungen und/oder Jod und/oder einer jodhaltigen Verbindung und/oder einer Mischung verschiedener jodhaltiger Verbindungen durchgeführt wird.

7. Verfahren nach einem der Ansprüche 1 bis 6, dadurch gekennzeichnet, dass die Rauchgasreinigung eine mehrstufige nasse Rauchgaswäsche mit mindestens einer stark sauren und/oder mindestens einer schwach sauren und/oder alkalischen Waschstufe umfasst.

8. Verfahren nach einem der Ansprüche 1 bis 7, dadurch gekennzeichnet, dass die Rauchgasreinigung mindestens eine trockene oder quasitrockene, auf Adsorption basierende Reinigungsstufe umfasst, insbesondere unter Verwendung von elektrostatischen oder filternden Staubabscheidern.

9. Verfahren nach Anspruch 8, dadurch gekennzeichnet, dass die mit Quecksilber beladenen Flugstäube aus gegebenenfalls vorhandenen Staubabscheidern zwecks Quecksilberentfrachtung thermisch nachbehandelt wird, insbesondere in einer auf Temperaturen von mindestens 200°C beheizten Drehtrommel.

10. Verfahren nach einem der Ansprüche 1 bis 9, dadurch gekennzeichnet, dass der Quecksilbergehalt des Rauchgases, insbesondere der Gehalt an metallischem Quecksilber, nach der Rauchgasreinigung kontinuierlich gemessen wird und anhand des gemessenen Quecksilbergehalts die Menge an zugeführtem Brom und/oder bromhaltigen Verbindungen sowie gegebenenfalls Schwefel und/oder schwefelhaltigen Verbindungen geregelt wird.

Es folgen 9 Blatt Zeichnungen

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Anhängende Zeichnungen

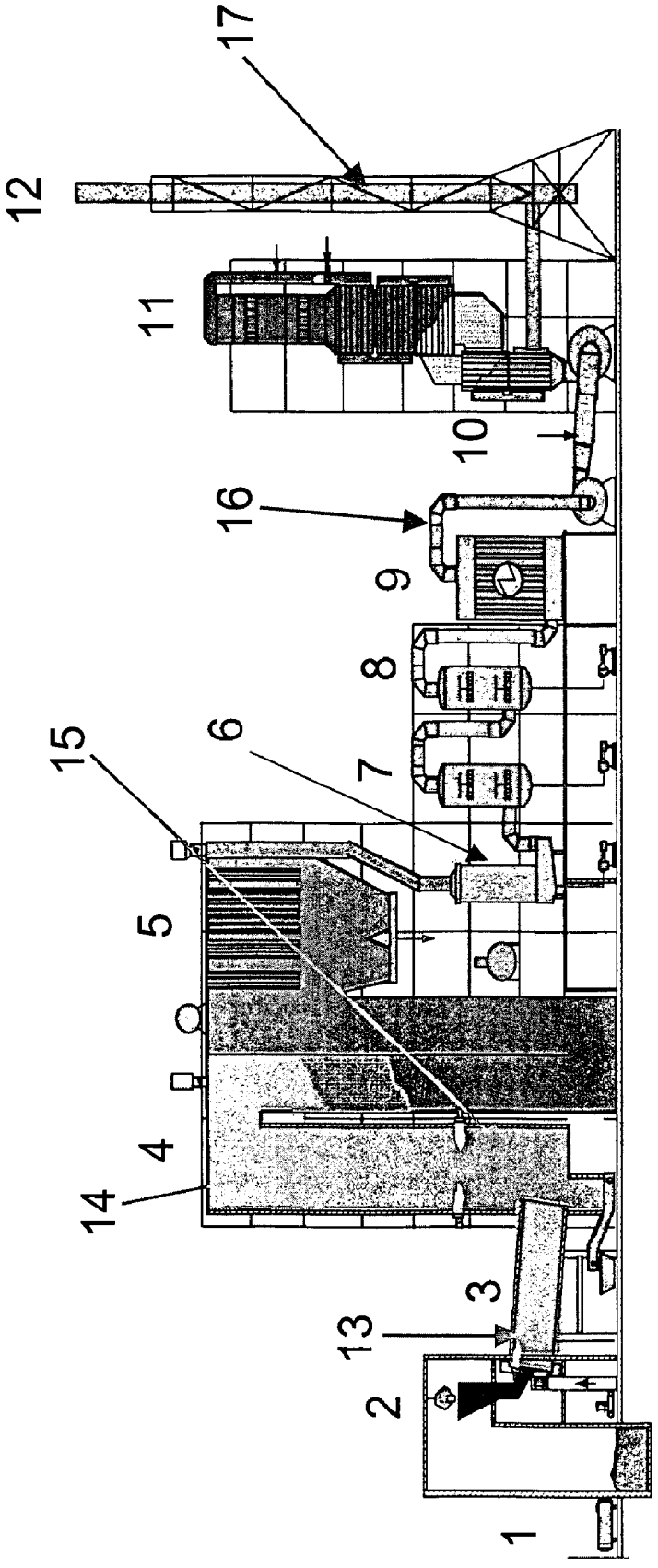


Fig. 1

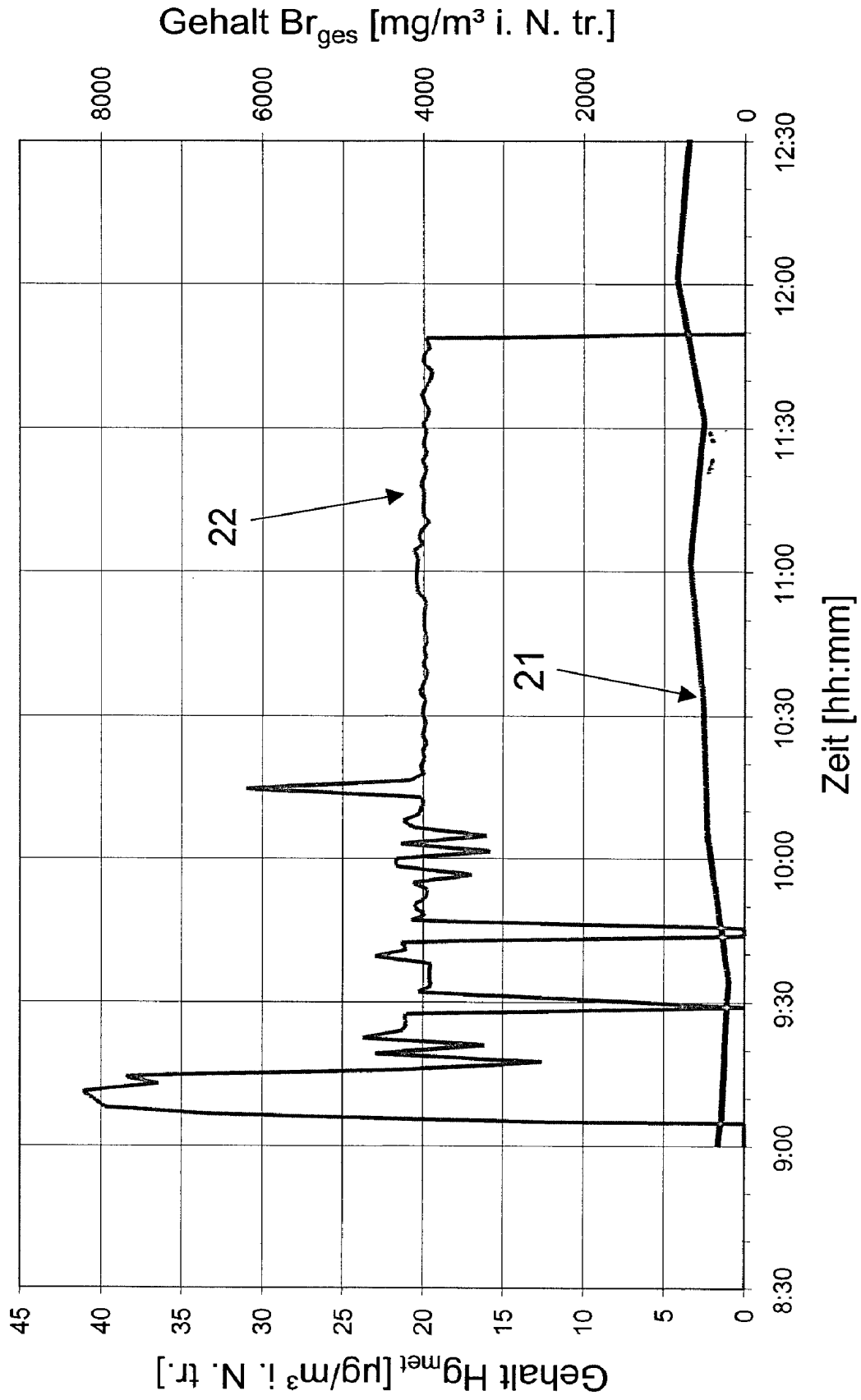

Gehalt Br_{ges} [mg/m³ i. N. tr.]
8000
6000
4000
2000
0
22
21
Gehalt Hg_{met} [µg/m³ i. N. tr.]
45
40
35
30
25
20
15
10
5
0
8:30
9:00
9:30
10:00
10:30
11:00
11:30
12:00
12:30
Zeit [hh:mm]

Fig. 2

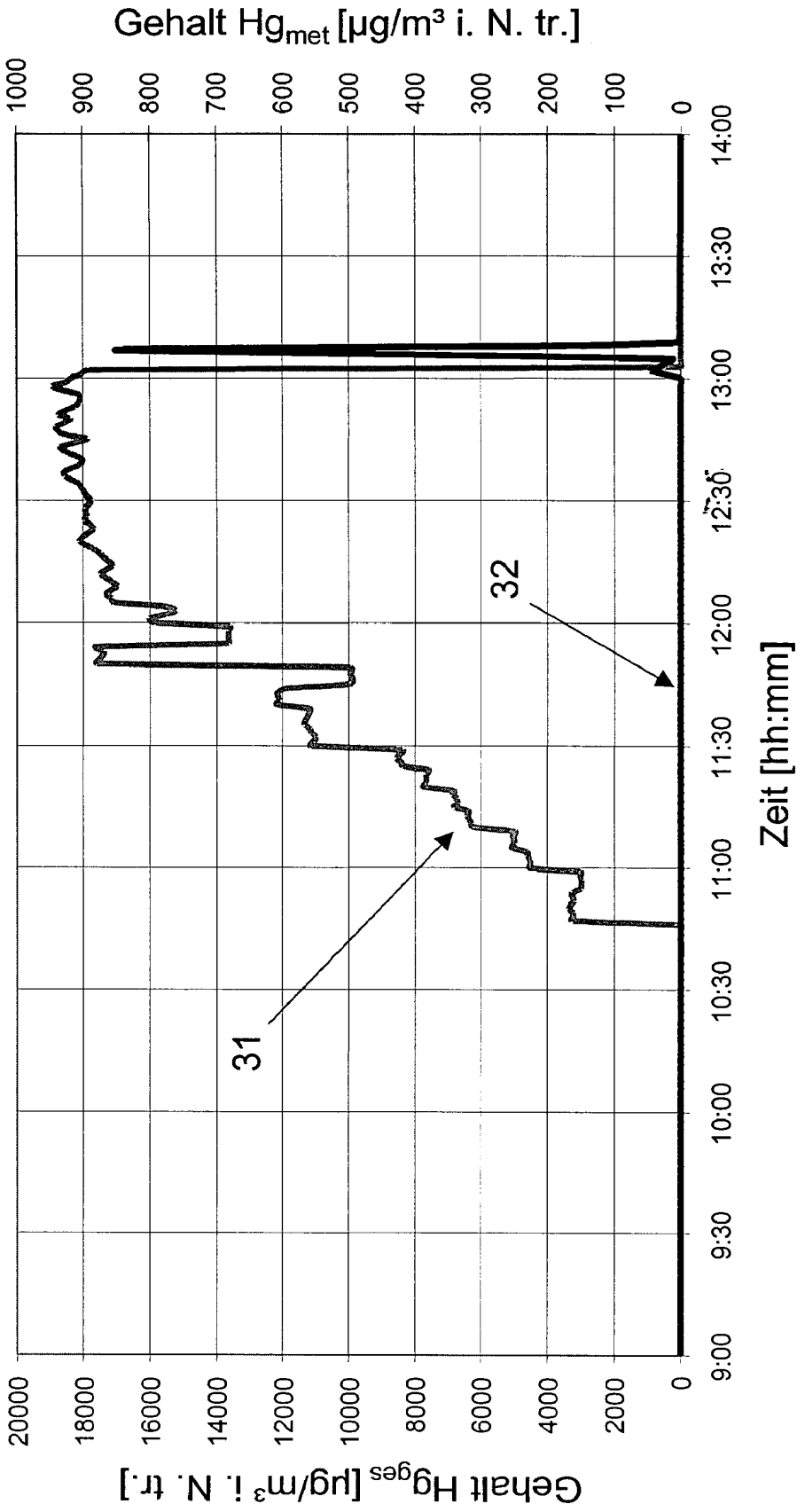

Gehalt Hg_{met} [µg/m³ i. N. tr.]
1000
900
800
700
600
500
400
300
200
100
0
14:00
13:30
13:00
12:30
12:00
11:30
11:00
10:30
10:00
9:30
9:00
Zeit [hh:mm]
32
31
Gehalt Hg_{ges} [µg/m³ i. N. tr.]
20000
18000
16000
14000
12000
10000
8000
6000
4000
2000
0

Fig. 3

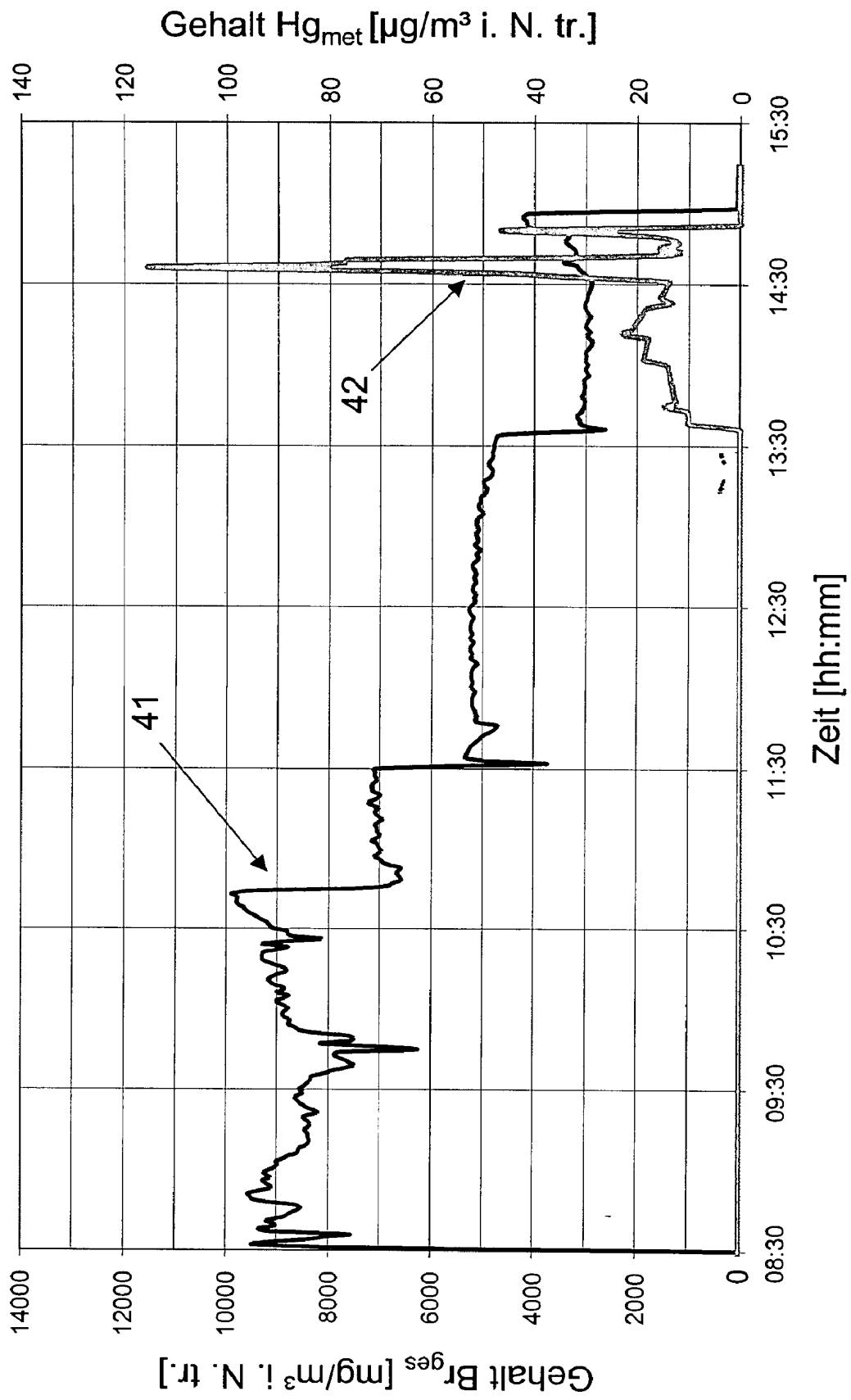

Gehalt Hg_{met} [µg/m³ i. N. tr.]
140
120
100
80
60
40
20
0
15:30
14:30
13:30
12:30
11:30
10:30
09:30
08:30
Zeit [hh:mm]
14000
12000
10000
8000
6000
4000
2000
0
Gehalt Br_{ges} [mg/m³ i. N. tr.]
41
42

Fig. 4

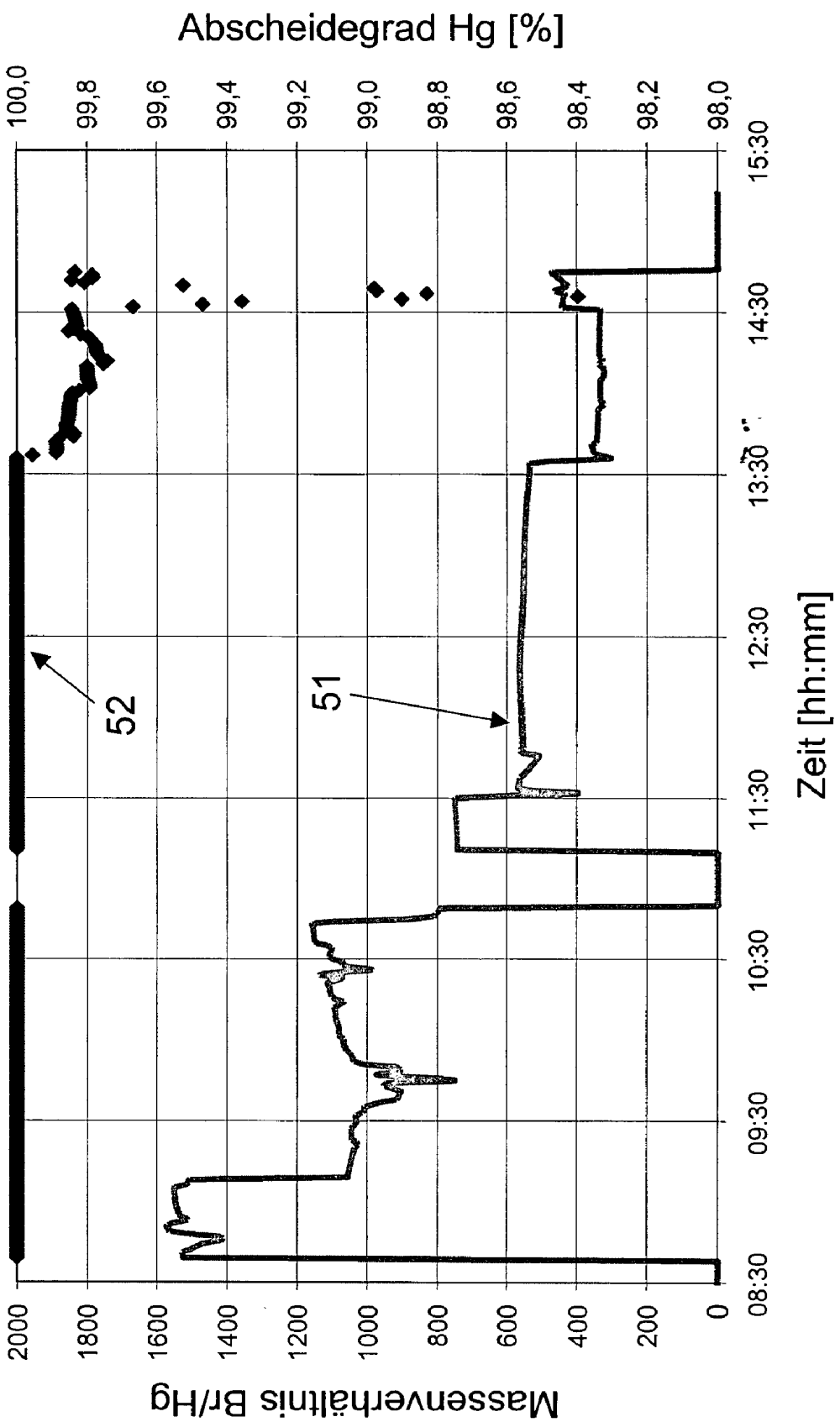

Abscheidegrad Hg [%]
100,0
99,8
99,6
99,4
99,2
99,0
98,8
98,6
98,4
98,2
98,0
2000
1800
1600
1400
1200
1000
800
600
400
200
0
08:30
09:30
10:30
11:30
12:30
13:30
14:30
15:30
Zeit [hh:mm]
Massenverhältnis Br/Hg
51
52

Fig. 5

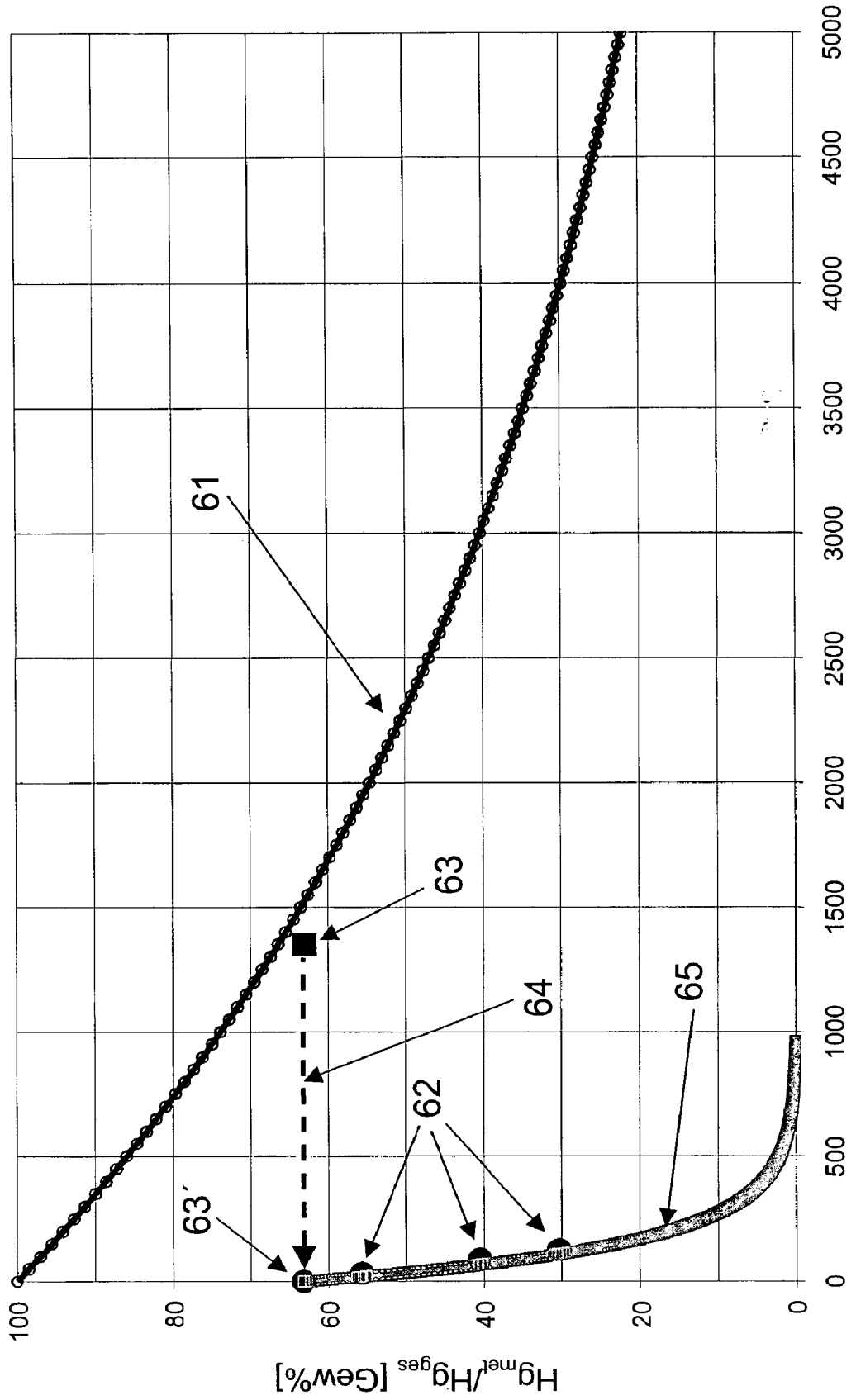

Hg_{met}/Hg_{ges} [Gew%]
100
80
60
40
20
0
0
500
1000
1500
2000
2500
3000
3500
4000
4500
5000
Gehalt Br_{ges} bzw. Cl_{ges} [mg/m³ i. N. tr.]
61
63
64
65
62
63´

Fig. 6

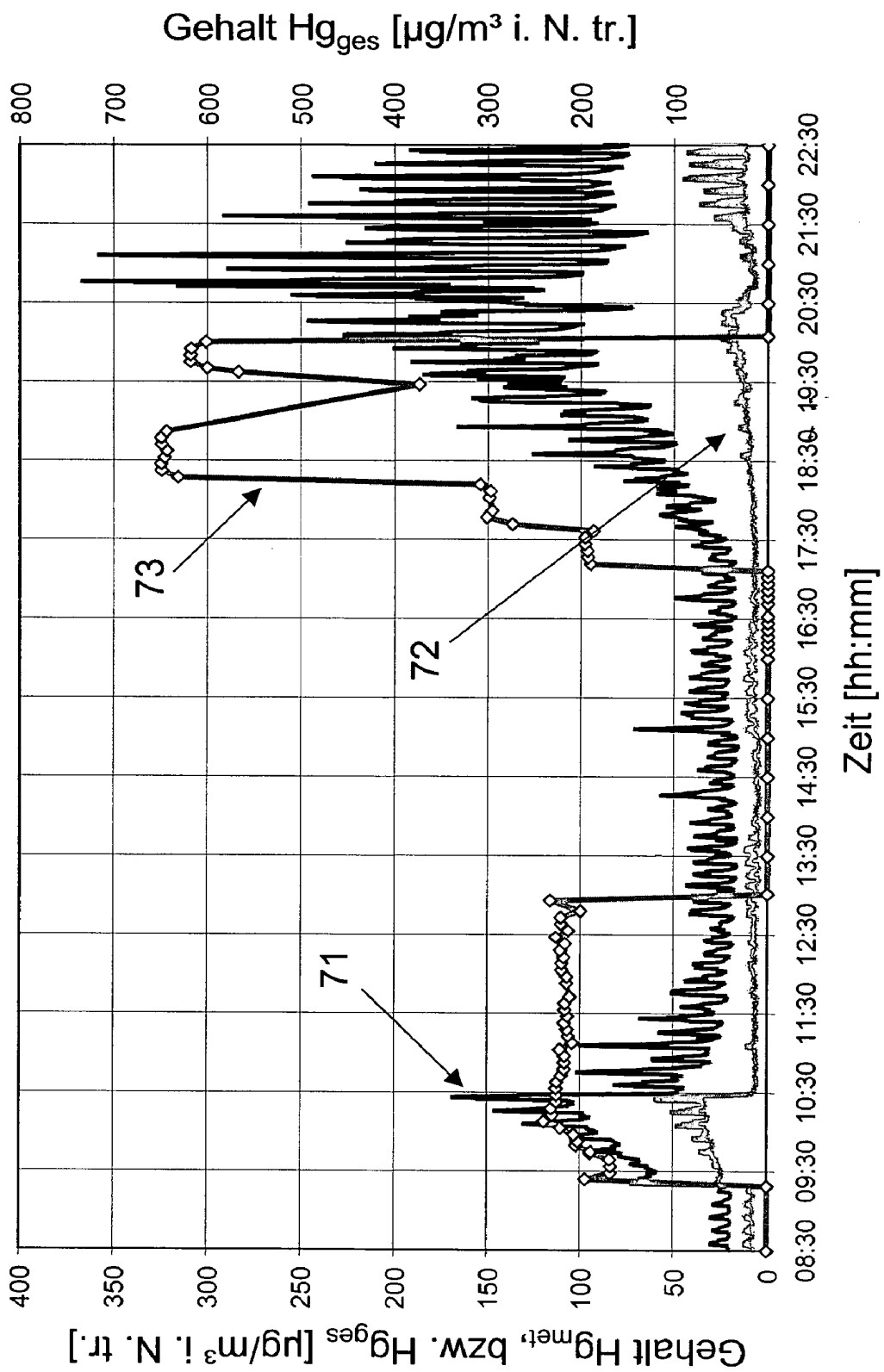

Gehalt Hg_{ges} [µg/m³ i. N. tr.]
800
700
600
500
400
300
200
100
08:30
09:30
10:30
11:30
12:30
13:30
14:30
15:30
16:30
17:30
18:30
19:30
20:30
21:30
22:30
Zeit [hh:mm]
Gehalt Hg_{met}, bzw. Hg_{ges} [µg/m³ i. N. tr.]
400
350
300
250
200
150
100
50
0
71
72
73

Fig. 7

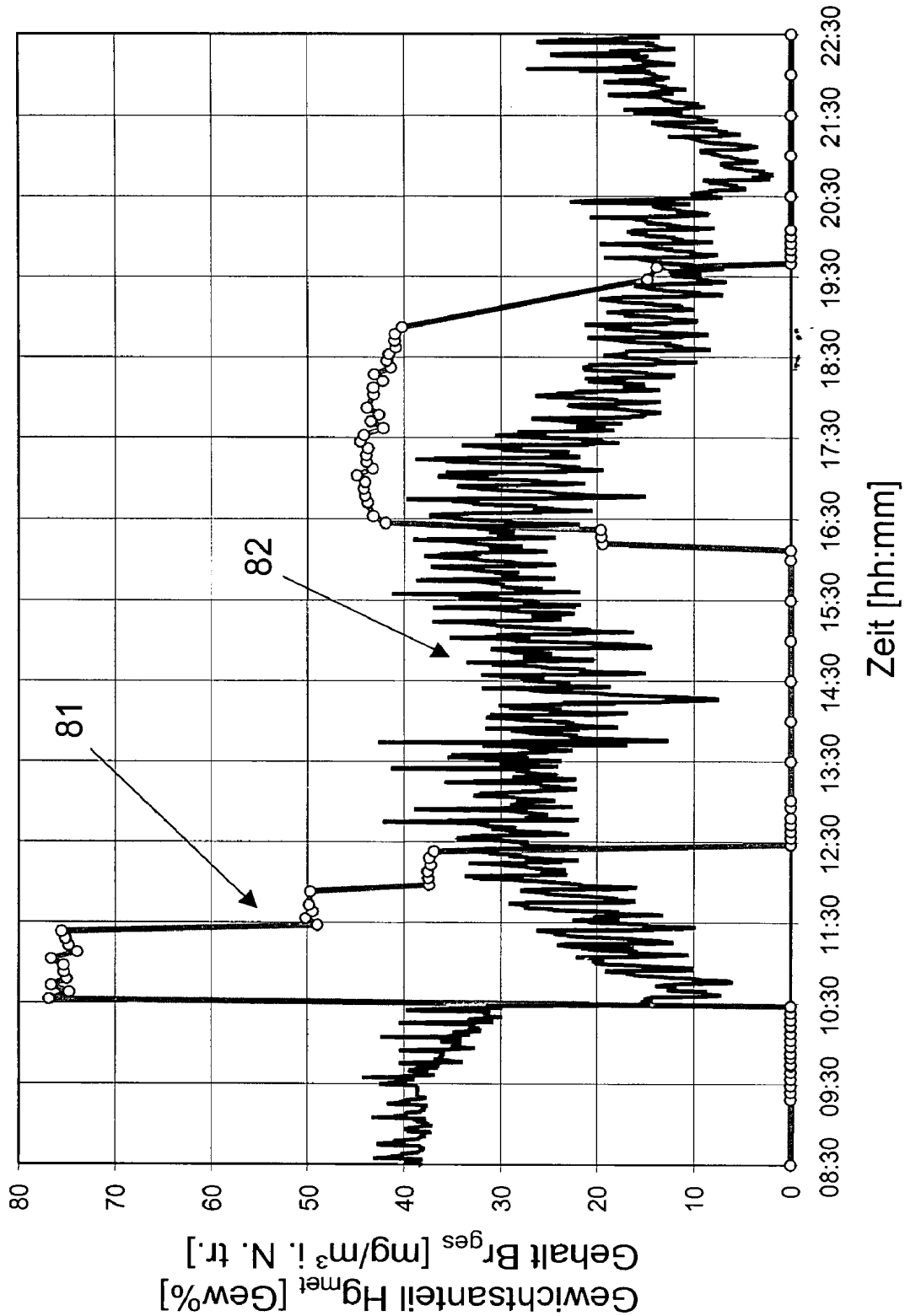

Gehalt Br_{ges} [mg/m³ i. N. tr.]
Gewichtsanteil Hg_{met} [Gew%]
0
10
20
30
40
50
60
70
80
08:30
09:30
10:30
11:30
12:30
13:30
14:30
15:30
16:30
17:30
18:30
19:30
20:30
21:30
22:30
Zeit [hh:mm]
81
82

Fig. 8

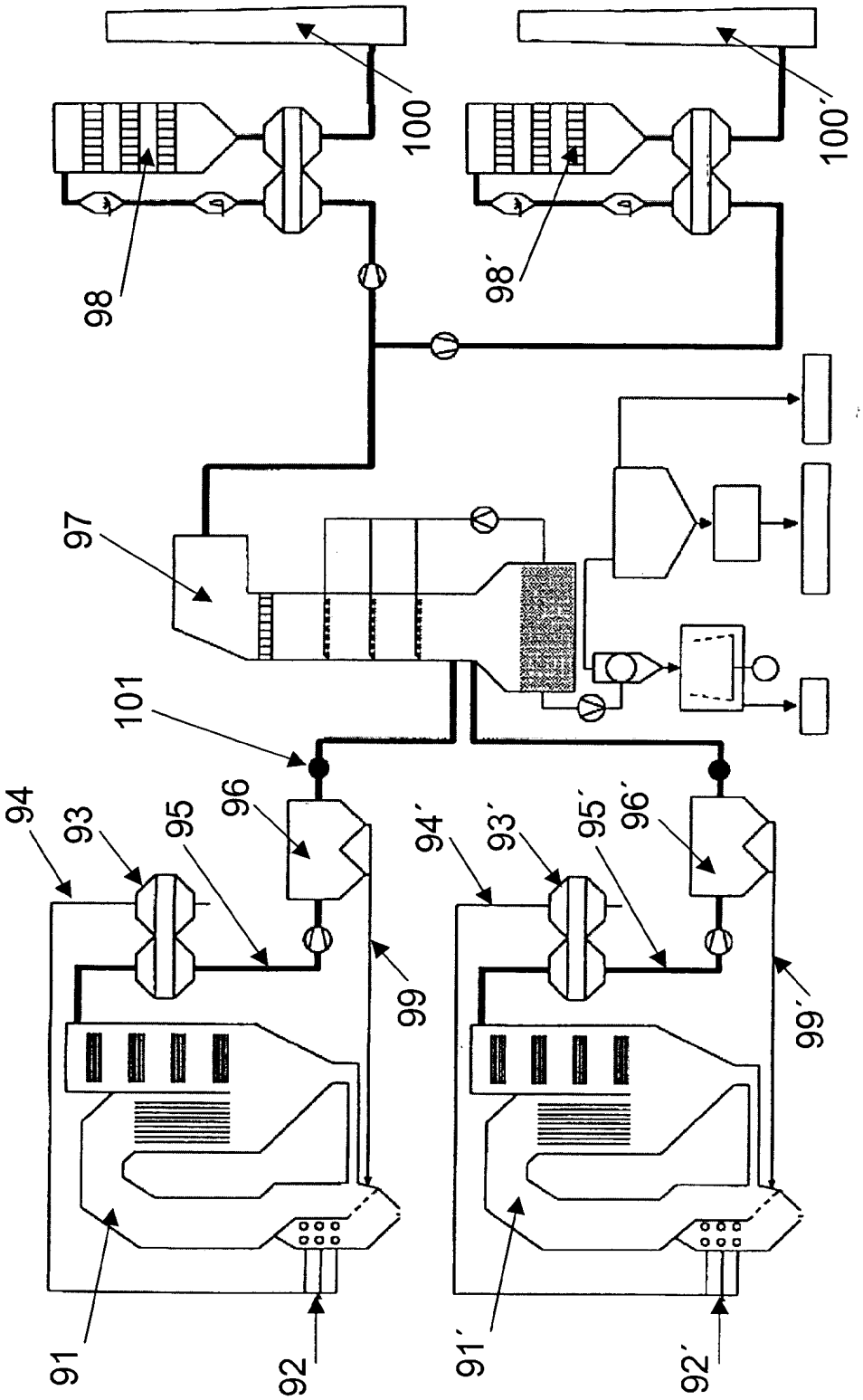


Fig. 9

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(54) **A process for removal of mercury vapor and vapor of chlorodibenzodioxins and -furans from a stream of hot flue gas.**

(57) Mercury vapor and/or vapor of chlorodibenzodioxins and/or vapor of chlorodibenzofurans are removed from a stream of hot flue gas together with acidic components of the flue gas in a spray absorption process. The absorbent used in said process is an aqueous liquid which besides alkaline components contains suspended activated carbon.

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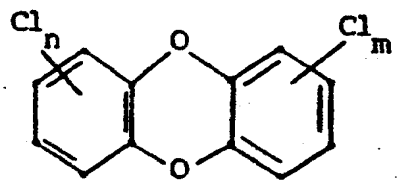
A process for removal of mercury vapor and vapor of chlorodibenzo-dioxins and -furans from a stream of hot flue gas.

FIELD OF THE INVENTION.

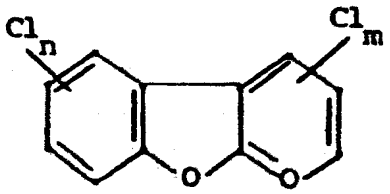
It is a well recognized fact that flue gases often contain mercury vapor in such amounts that their release into the atmosphere is regarded as being hazardous. This especially applies to flue gases from incineration plants where domestic refuse is incinerated but also flue gas formed by combustion of certain coals may contain mercury vapor in such amounts that it is highly desirable to remove a substantial part of said mercury vapor before releasing the flue gas into the atmosphere.

In the present specification and the attached claims the term mercury vapor means elementary mercury as well as mercury-containing chemical compounds in the vapor phase.

In recent years a growing concern has arisen as to the presence in flue gases of the toxic polychlorodibenzodioxines (PCDD) and polychlorodibenzofurans (PCDF). Said two series of compounds may be represented by the following formulae



PCDD



PCDF

wherein n and m each independently is an integer from 0 through 4 provided that n + m is at least 2.

PCDD and PCDF are present in flue gases from incineration plants and in minor proportion also in the

flue gases from power plants. In flue gases these compounds are primarily present in vapor phase and in the present specification the above abbreviations denote the compounds as vapors.

The extremely high toxicity of certain of these compounds makes it desirable to avoid their release into the atmosphere.

DESCRIPTION OF PRIOR ART.

Numerous methods have been suggested for removing or recovering mercury from gases. However, the majority of the prior art processes have been created with the purpose of removing mercury from relative small amounts of gas having high mercury concentration. These processes are not suitable for cleaning flue gas since costs of chemicals would be prohibitive or operation would be impracticable in connection with large volumes of flue gas.

Processes for removing mercury from air of relatively low mercury content have also been suggested. Such a process is disclosed in published European Patent Application No. 1.456. (Akzo N.V.). Said process which is described as particularly suitable for the removal of mercury from air which is vented from buildings, is based on the principle that mercury vapor is absorbed as mercury chloride when passing a bed of activated carbon having a specific chlorine content. According to the specification to said European application high moisture content of the gas from which mercury is to be removed, should be avoided since the effectiveness of the activated carbon is reduced thereby. From said specification it also appears that activated carbon used in a stationary bed without chlorine is unsatisfactory as absorbent for mercury and has a very low capacity for that purpose.

The process of said European application seems unsuitable for treating flue gas since it would require the total amount of flue gas to be passed through a bed of activated carbon to which gaseous chlorine is added, which obviously involves the risk that a surplus of chlorine may be entrained with the flue gas to the atmosphere.

A process for removing mercury vapor from a hot hydrogene chloride-containing flue gas is disclosed in published European patent application no. 13.567, (Svenska Fläktfabriken). According to said process the gas which contains hydrogen chloride and minor amounts of mercury vapor, is contacted with powdered calcium hydroxide, preferably in a fluidized bed. The hydrogen chloride in the gas reacts with the calcium hydroxide to form calcium chloride which apparently is essential to the removal of mercury. Said process is restricted to gases having a substantial contents of hydrogen chloride. This condition will often be fulfilled by the flue gas from a waste incineration plant while flue gasses from other sources may be deficient in that respect.

US patent specification 4.273.747 (Rasmussen) discloses removal of mercury from hot waste gasses by atomizing an aqueous liquid into the waste gasses in the presence of fly ash suspended in the gas and subsequently separating the fly ash together with a substantial part of the mercury originally present as vapor. It is essential that by said treatment the gas stream is cooled from a temperature of at least 200°C to a temperature below 160°C. The aqueous liquid may be just water or it may be an aqueous solution or suspension of an alkaline compound, preferably calcium hydroxide.

Obviously said method will not be suitable in case it is not acceptable to cool the gas to the

extent required or if the amount of fly ash is insufficient due to the use of a preceding fly ash separation. Even when the conditions as to fly ash contents of the flue gas and cooling are satisfied it would in certain applications be desired to increase the efficiency of the removal of mercury vapor in said process.

Efforts to reduce the PCDD and PCDF levels in flue gas have hitherto mainly concentrated on thermal destruction. A reliable thermal destruction is, however, difficult to establish, since a very high temperature is required to ensure a complete destruction.

According to a paper of A.J. Teller and J. D. Lauber : "Control of Dioxin Emissions from Incineration" presented at the 76th annual meeting of the Air Pollution Control Association, Atlanta, Georgia, June 19 - 24, 1983, dioxin theoretic estimations indicate that emission may be reduced by condensing the dioxin thereof. However, A. Nottrodt et al: "Emissionen von polychlorierten Dibenzodioxinen und polychlorierten Dibenzofuranen aus Abfallverbrennungsanlagen", Müll und Abfall 11/84, p. 313-327 (Erich Schmidt Verlag GmbH Berlin, Bielefeld, München) state results showing high vapor phase concentrations after several wet scrubbings with flue gas temperatures lower than those obtained in the theoretic estimations made in said paper by Teller and Lauber.

Consequently a need excists for efficient measures for removing not only mercury vapor but also PCDD and PDCF vapors from flue gases.

SUMMARY OF THE INVENTION.

The present invention is directed to a novel process for efficient removal of mercury vapor and PCDD

and PCDF from a stream of hot flue gas, which process provides a simultaneous removal of acidic components of the flue gas.

It has turned out that incorporation of a relatively small amount of activated carbon in an alkaline feed suspension to be spraydried in a spray absorption process enables a surprising improvement of the ability of the process to remove mercury, PCDD and PCDF.

Consequently the process according to the invention is characterized in that an aqueous liquid which besides alkaline components comprises suspended activated carbon is spray dried in the flue gas which subsequently is freed from the resulting particulate material.

The amount of activated carbon to be used depends on the amount of mercury vapor as well as PCDD and PCDF in the flue gas, on the limits for emission which it is intended to meet, on the extent to which cooling of the gas is permissible, on the proportion of fly ash and the character thereof, etc.

Usually the amount of activated carbon will correspond to at least 5 mg per Nm^3 flue gas to be treated. For economic reasons the amount will usually not exceed 500 mg per Nm^3 flue gas, peferrably not 200 mg per Nm^3.

Since the prior art describes activated carbon as not being very suitable for absorption of mercury vapor when no chlorine is present and since high moisture conditions are described as harmful to mercury sorption by means of activated carbon together with chlorine it could not be predicted that a very efficient mercury removal is obtained by the very simple measures prescribed by the present process. It is also surprising that the rather small additions of activatet carbon to

the aqueous liquid to be atomized, which come into consideration by the present process, enables a reduction of PCDD and PCDF levels to extremely low values.

The preferred alkaline component for the simultaneous removal of acidic components of the gas is calcium hydroxide which by chemical reaction absorbs e.g. sulphur oxides and hydrogen chloride. However, it is to be observed that by the process of the invention presence of hydrogen chloride and consequently production of calcium chloride is not necessary to achieve an efficient removal of mercury vapor.

The fact that in the present process the activated carbon is initially present in aqueous suspension in the atomized droplets and subsequently is embedded in the particulate material comprising the reaction products of e.g. calcium hydroxide and the acidic components of the gas, apparently does not have any substantial adverse influence on the absorption ability of the activated carbon, as would have been expected.

The process of the present invention may be performed using those measures which are conventional in spray absorption processes for desulfurization of flue gases, e.g. as described in US patent 4,279,873 (Felsvang et al), incorporated herein by reference.

In the typical embodiment the process of the invention is performed on a stream of flue gas the fly ash of which has previously been removed in a particle collector, e.g. an electrostatic precipitator. However, the absorption of mercury vapor by activated carbon may be used as a supplement to the absorption by means of fly ash as described in the above cited US specification 4,273,747, in which case at least a portion of the fly ash present in the flue gas stream remains in said stream during the spray absorption process and is re-

covered together with the particulate material formed by the spray absorption process.

It is also possible to add fly ash to the liquid to be atomized, thereby utilizing the sorption ability of the fly ash to reduce the requirement for activated carbon.

Also other absorbents such as silicates may be added to the liquid to be atomized to minimize the requirement as to activated carbon.

As indicated above the amount of activated carbon necessary to achieve a substantial reduction of the concentration of mercury vapor and PCDD and PCDF in the flue gas is surprisingly small. The actual amounts to be used in each individual case may easily be fixed by a person skilled in the art on the basis of a few tests.

It is an important feature of the invention that by very simple measures a very flexible process is obtained since even flue gases with substantially varying levels of Hg, PCDD and PCDF may be coped with just by changing the amount of activate carbon added to the liquid to be atomized.

The particulate material formed in the spray absorption process and typically comprising calcium sulfite, calcium sulfate, calcium hydroxide and activated carbon and possibly fly ash is preferably separated from the flue gas in a baghouse which enables a prolonged contact between the particulate material and the flue gas.

In the following the process according to the invention will be further illustrated by means of examples.

In these examples analysis was performed on flue gas withdrawn by means of a heated probe with glass lining and equipped with a glass wool filter. The mercury was collected in two impinger flasks in series

containing an aqueous 5% potassium permanganate - nitric acid solution. Mercury was analyzed by flameless atomic absorption using $NaBH_4$ as a reductant (MHS-20, Mercury/ Hydride System by Perkin Elmer, ref. 338 - A2-M 294/ 12.79).

It is to be observed that methods previously used for determination of Hg in flue gases have not always been accurate and have often indicated values lower than the actual ones. Consequently the reults obtained as to removal of Hg vapor in the following examples may not be comparable with prior art results determined by means of less accurate methods of analysis.

EXAMPLE 1

Eight test runs were performed using a partial stream of the flue gas from an incineration plant wherein domestic waste was incinerated.

The spray absorption process was performed in a spray absorber the chamber of which comprised a cylindrical portion having a diameter of 1.2 m and a hight of 0.8 m, and a conical bottom portion the cone angle of which was 60°. The spray absorber was provided with a rotary atomizer. Downstream of the spray absorber particle separation was performed using a baghouse.

An aqueous suspension of slaked lime or an aqueous solution of sodium carbonate (solids content of the suspension or solution app. 5%) was used as feed for the atomizer.

The temperature of the flue gas when led to the spray absorber was 225-300°C and the amount of fly ash therein was 0.5-1.0 g/Nm^3 in run nos. 1-5 whereas run nos. 6-8 were performed using flue gas with substantially no fly ash. The flue gas was supplied in an amount of 300 kg/h.

The concentration of mercury vapor was measured upstream of the spray absorber, between the spray ab-

sorber and the baghouse filter and downstream of the baghouse filter.

Run nos. 1 and 7 were performed for comparison and no activated carbon was added to the feed suspension of slaked lime or feed solution of sodium carbonate. In runs 2 through 6 and in run 8 activated carbon was added to said feed solution in an amount of app. 2% by weight of the solids content, corresponding to 0,04-0,05 g/Nm^3 of flue gas.

The following results were obtained:

	Run no. 1	2	3	4	5	6	7	8
Upstream of spray absorber								
Hg vapor, µg/Nm^3	241	290	725	517	314	423	213	405
Absorbent	Slaked lime	Slaked lime + activated carbon	slaked lime + activated carbon	slaked lime + activated carbon	slaked lime + activated carbon	slaked lime + activated carbon	Na_2CO_3	Na_2CO_3 + activated carbon
Between spray absorber and baghouse								
Gas temperature, °C	130	130	100	160	175	165	130	130
Hg vapor, µg/Nm^3	77	46	not analysed	not analysed	not analysed	not analysed	not analysed	not analysed
Downstream the baghouse								
Hg vapor, µg/Nm^3	30	20	11	42	63	80	96	47
% removal	87.6	93	98.9	92.9	79.9	81.1	54.9	88.4

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The above results clearly indicate that activated carbon acts as a very efficient absorbent for mercury vapor when utilized in the process of the present invention.

EXAMPLE 2

Two further test runs was made in the same plant and under similar conditions as described in example 1, except that the flue gas temperature varied between 280 and 300°C.

The results obtained are listed in the following table:

	Run No. 9	Run No. 10
Upstream of spray absorber		
PCDD[1] +PCDF, µg/Nm^3	0.38	0.53
Absorbent	slaked lime	slaked lime + activated carbon
Between spray absorber and baghouse		
Gas temperature, °C	150	150
PCDD[1] +PCDF, µg/Nm^3	not analysed	0.28
Downstream the baghouse		
PCDD[1] +PCDF, µg/Nm^3	0.047	0.01
% removal	87.8	98.1

[1] The PCDD contents were approximately 35-45% of the total contents of PCDD and PCDF.

CLAIMS

1. A process for removal of mercury vapor and/or vapor of chlorodibenzodioxins and/or vapor of chlorodibenzofurans from a stream of hot flue gas combined with a simultaneous removal of acidic components of the flue gas, characterized in that an aqueous liquid, which besides alkaline components comprises suspended activated carbon is spray dried in the flue gas which subsequently is freed from the resulting particulate material.

2. The process of claim 1, wherein the amount of activated carbon in said aqueous liquid amounts to 5-500 mg per Nm^3 flue gas being treated.

3. The process of claim 1, wherein the amount of activated carbon in said aqueous liquid amounts to 5-200 mg per Nm^3 flue gas being treated.

4. The process of claim 1, wherein said alkaline components comprise calcium hydroxide in a quantity at least sufficient to neutralize a substantial part of the acidic components, such as sulfuroxides and hydrogen chloride, of the flue gas.

5. The process of claim 1, wherein said particulate material is recovered in a baghouse.

6. The process of claim 1, wherein at least a portion of any fly ash present in the flue gas stream remains in said stream during the spray absorption process and is recovered together with the particulate material formed by the spray absorption process.

7. The process of claim 1, wherein at least a portion of any fly ash present in the flue gas stream is collected from said stream before said aqueous liquid is spray dried therein, and at least a part of the collected fly ash is suspended in said aqueous liquid before spraying thereof.

European Patent Office

EUROPEAN SEARCH REPORT

Application number
EP 86 30 5054 0208490

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
A	US-A-4 049 399 (TELLER ENVIRONMENTAL SYSTEMS)		B 01 D 53/34
A	US-A-2 886 131 (J.R. CONLISK)		
D,A	US-A-4 273 747 (E. RASMUSSEN)		
A	US-A-4 246 242 (C.H.S. BUTLER)		TECHNICAL FIELDS SEARCHED (Int. Cl.4)
A	CHEMICAL ABSTRACTS, vol. 88, no. 2, 9th January 1978, page 125, abstract no. 9181j, Columbus, Ohio, US; T. HALMOE: "Micropollutants from an incineration plant lower than expected", & FAST AVFALL 1977, (1), 29-31		B 01 D

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	26-08-1986	PYFFEROEN K.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
Y : particularly relevant if combined with another document of the same category
A : technological background
O : non-written disclosure
P : intermediate document

T : theory or principle underlying the invention
E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document

EPO Form 1503 03.82

⑤1 Int・Cl.　⑤2日本分類
B 01 d 53／02　13(9)F 2
B 01 d 53／04　13(7)B 62
B 01 d 53／34　13(7)A 11

⑲日本国特許庁

特許公報

⑪特許出願公告
昭49—43197
㊹公告　昭和49年(1974)11月19日
発明の数　1
（全3頁）

1

⑤4ガス中の水銀除去方法

㉑特　願　昭45—124108
㉒出　願　昭45（1970）12月29日
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発明の詳細な説明

本発明はガス中に含まれる微量の水銀を除去する方法に関する。

例えば水銀法アルカリ塩電解で得られる水素ガス中には水銀を含み、これは微量でも有機化合物の接触還元等に水素を使用する際、触媒毒となるため、あらかじめ実質的に除去しておくことが要求される。このような場合、水銀の除去方法としては水素を液体アンモニアの真空蒸発により−40℃に冷却してある竪型の管型冷却器に送り冷却し、水銀を微粒子に凝縮して除く深冷凝縮法、或いは水銀蒸気を含むガスを3～5kg／cm²の圧力の下に−30℃以下に冷却して除去する方法があるが、いずれも気体冷却のため膨大な冷凍設備を必要とする。

さらにモレキユラーシーブ又は活性炭により選択的に吸着させる方法、あるいは水銀を含むガスを活性炭または酸化アルミニウム上で硫化水銀と硫黄の混合物と接触せしめる方法があるが、前者は比較的能率の悪い欠点があり又後者は微量な水

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銀分が完全には吸着されない欠点がある。

又化学的方法として塩素水と水銀含有水素ガスを接触させて、水銀塩化物として除去する方法が知られているが操作時の調節が難かしく余分の塩素によりガスを再汚染する欠点がある。

又オゾン処理による除去方法も考案されているが後処理の静電分離、又は機械的ろ過に難点がある。さらにヨウ素を活性炭に吸着せしめた担体で除去方法があるが、ヨウ素が逸散する為に連続使用の点に難点があつた。

本発明者らは以上の問題点を解決するために検討をかさねた結果、水銀との反応性に富んだ遊離ヨウ素を含有しないヨウ素化合物および還元剤を利用してきわめて効果的にガス中の水銀を除去することに成功したものである。すなわち本発明はガス中の水銀を除去するに当り遊離ヨウ素を含有しないリチウム、ナトリウム、カリウム、カルシウム、マグネシウム、亜鉛、アルミニウム、鉄、ニツケル、コバルトより選ばれた金属ヨウ化物、あるいは一般式R_4NI（但しRはHまたはC_1～C_4のアルキル基）で示される化合物を炭素系担体、ゼオライト、シリカゲルのいづれかに吸着させた吸着剤担体に無機還元剤を1種または2種以上共存せしめて水銀を含むガスを接触させることを特徴とするガス中の水銀除去方法である。

このようなヨウ素化合物としてはヨウ化リチウム、ヨウ化ナトリウム、ヨウ化カリウム、ヨウ化カルシウム、ヨウ化マグネシウム、ヨウ化亜鉛、ヨウ化アルミニウム、ヨウ化鉄、ヨウ化ニツケル、ヨウ化コバルト、ヨウ化テトラエチルアンモニウムなどが挙げられる。また共存させる無機還元剤としてはヒドラジニウム塩、ヒドロキシルアンモニウム塩、亜リン酸及びその塩、チオ硫酸、次亜硫酸、亜硫酸及びその塩、第1錫化合物などが挙げられる。

また本発明に用いられる吸着剤担体の粒度は約6～100メツシユが適当である。

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本発明方法を実施するためにはあらかじめ該ヨウ素化合物を水に溶解させて、吸着剤担体の充填層に流下させることにより充分担体表面にヨウ素化合物を吸着せしめるか、あるいは該ヨウ素化合物を溶解させたアルコール溶媒に吸着剤担体に加えて充分接触攪拌後、アルコールを逸散せしめて担体表面にヨウ素化合物を吸着せしめる。さらに無機還元剤を共存させるために、その水溶液を同様にして担体表面に吸着させた後水銀を含むガスを通ずればよい。本発明方法は水素以外のガス、たとえば空気、窒素、塩酸ガスなどにも適用しうるものである。またガスの種類によつて耐用性が劣化を来したり、あるいはガス中の水銀の形態は通常水銀蒸気であるがガスの種類によつてそのごく一部が酸化水銀のごとき水銀化合物になつていることもあり、本発明法のごとく還元剤を共存せしめることにより効果的に水銀を除去しうるものである。

なおヨウ素化合物の吸着量はヨウ素分として吸着剤担体の重量の２～２５重量%程度、無機還元剤の吸着量は担体重量の0.１～５重量%程度が適当である。

このようにして水銀含量約１０００～40000 μg／m³（μgはmgの１０００分の１）のガスを空間速度２００～４０００ｌ／Ｈｒで充填層に流通せしめると、排ガス中の水銀は１０μg／m³以下に減少する。

ガス中におけるヨウ素化合物と水銀との反応機構は明らかではないが、最終的には吸着もしくは担持せしめたヨウ素化合物が水銀と反応してこれを捕促するものと思われる。

又還元剤を共存させることによる反応機構は明らかではないが最終的には担体に存在するヨウ素化合物が水銀と反応してこれを捕捉すると同時に共存せしめた還元剤により耐用性がよくなりさらに酸化水銀の如き水銀化合物も還元されてより効果的に水銀を除去しうるものと思われる。

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なお還元剤としては種々知られているが本発明方法に用いられる還元剤はこれらのうちで特に還元力にとみ、又耐用性を助長することが大で、実用化に適しているものである。

参考例　１

６～２０メッシュの活性炭４０ｇを内径２０㎜のガラス管に充填し水銀含量１２０００μg／m³の水素ガスを空間速度１５００ｌ／Ｈｒで流通せしめたところ排出ガス中の水銀濃度は１時間後で１５０μg／m³、２０時間後で３２００μg／m³９０時間後で１１７００μg／m³であつた。

参考例　２

２０～３０メッシュのゼオライト３０ｇを内径２０㎜のガラス管に充填し水銀含量９７００μg／m³の水素ガスを空間速度１０００ｌ／Ｈｒで流通せしめたところ排出ガス中の水銀濃度は１時間後で９６００μg／m³であつた。

実施例　１

１０～４０メッシュの活性炭４０ｇを内径２０㎜のガラス管に充填しあらかじめヨウ化ナトリウム１０ｇを溶解しているエタノール１５０mℓを流下せしめて担体に充分吸着せしめたのち風乾し、溶媒を充分除去する。さらに硫酸第一錫２ｇを含む水溶液５０mℓを流下せしめて硫酸第一錫を共存せしめた後、水銀含量２５０μg／m³の塩酸ガスを空間速度１５００ｌ／Ｈｒで流通せしめたところ排出ガス中の水銀濃度は１０時間後で２μg／m³、９０時間後で３μg／m³、２５０時間後で５μg／m³、５００時間後で９μg／m³であつた。

実施例　２～４

実施例１と同様の方法でヨウ素化合物、担体および還元剤の種類をかえた実験結果を下表に示す。

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実施例	ヨウ素化合物	溶媒	担体			還元剤		対象ガス		ガス中の水銀濃度（μg／m³）		
点			種類	重量	ヨウ素化合物吸着量	種類	共存量	種類	流量空間速度	入口	出口	
				g	重量%		重量%		1／Hr		10時間後	250時間後	500時間後
2.	ヨウ化亜鉛	水	活性炭	40	約 4.5	塩化第一錫	約2.0	H_2	1500	14600	2	4	9
3.	ヨウ化アルミニウム	水	活性炭	40	約10.0	硫酸ヒドロキシルアミン＋硫酸第一錫	約2.0	H_2	1500	15000	3	4	10
4.	ヨウ化テトラエチルアンモニウム	水	ゼオライト	50	約 5.5	硫酸ヒドラジン	約1.5	H_2	600	9700	4	5	10

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㊼特許請求の範囲

1　ガス中の水銀を除去するにあたり遊離ヨウ素を含有しないリチウム、ナトリウム、カリウム、カルシウム、マグネシウム、亜鉛、アルミニウム、鉄、ニッケル、コバルトより選ばれた金属ヨウ化物、あるいは一般式R_4NI（但しRはHまたはC_1～C_4のアルキル基）で示される化合物を炭素系組体、ゼオライト、シリカゲルのいづれかに吸着させた吸着剤担体に無機還元剤を１種または２種以上共存せしめて、水銀を含むガスを接触させることを特徴とするガス中の水銀除去方法。

㊻引用文献

特　　公　昭４７－４４１５５
米国特許　３１９４６２９

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Page 114

WPI / Thomson

AN - 1974-86199V [50]

AP - JP19720097145 19720929

CPY - NIPS

DC - J01

DW - 197450

MC - J01-E01

PA - (NIPS) NIPPON SODA CO

PN - JP49053593 A 19740524 DW197450

XIC - B01D-053/46; B01D-053/64; B01J-020/20; C01B-031/00; C01B-031/08

AB - Activated carbon contg. >=1 of chlorides of Gp I and IIB metals is prepd. by immersing ordinary activated carbon in a soln. of the chlorides in water or an org. solvent. In an example, 50 pts activated carbon (20-50 mesh) was immersed in 200 pts aw. 10% ZnCl2 soln. for 30 min and dried at 105-110 degrees C for 10 hr after removing excess soln. Then, the activated carbon was packed in a glass tube and air contg. 10 mg Hg/m3 was passed through the tube at 1.1 l./min. Even after 10 hr, the activated carbon actively adsorbed Hg and the air thus treated contd. <0.005 mg Hg/m3.

ICAI- B01D53/64; B01J20/20; C01B31/08

ICCI- B01D53/46; B01J20/20; C01B31/00

IW - ACTIVATE CARBON CONTAIN MERCURY EXTRACT AIR GAS ADSORB

IWW - ACTIVATE CARBON CONTAIN MERCURY EXTRACT AIR GAS ADSORB

NC - 1

NPN - 1

PAW - (NIPS) NIPPON SODA CO

PD - 1974-05-24

TI - Activated carbon contg. chlorides for mercury extn. - from air or gases, with excellent adsorptivity

BNSDOCID: <XP_____2514926A_I_>

⑲ 日本国特許庁

公開特許公報

⑪特開昭 49－53593
㊸公開日 昭49.(1974)5.24
㉑特願昭 47-97145
㉒出願日 昭47.(1972)9.29
審査請求 未請求 (全3頁)

庁内整理番号	㊷日本分類
6646 41	14 E33
6415 4A	13(9)F2

(2,000円)

昭和47年9月29日

特許庁長官 三宅幸夫 殿

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5. 添付書類の目録
~~(イ) 出願審査請求書 1通~~
(ロ) 明細書 1通
~~(ハ) 図面 1通~~
(ニ) 委任状 1通
(ホ) 願書副本 1通

特許庁 47.9.29 出願第二課 北沢

明細書

1. 発明の名称
活性炭

2. 特許請求の範囲
週期律表の1族及び2族bの金属元素の塩化物から選ばれた1種又は2種以上の化合物を担持せしめることを特長とする活性炭。

3. 発明の詳細なる説明
本発明は、ガス中に存在する水銀に適用する新規な水銀吸着用活性炭及びその製造方法に係わるものである。

大気中に含まれる水銀は従来から労働衛生的立場より許容値(0.1 mg/m³)以下に管理することが必要であつたが、昨今の公害規制の強化に伴つて、環境衛生的立場からも次第に厳しい管理が必要とされるようになつてきている。同様に、又化学工業製品の原料となる各種の気体、例えば水素ガスなどに含まれる水銀に対しても厳しい規制と管理が必要になつている。

この様な大気中の水銀又は各種気体中の水銀の除去に関しては従来既にいくつかの方法が提案されている。例えば、高濃度の水銀に対しては、冷却して凝縮させる方法、又低濃度の水銀に対しては、酸化剤を含む溶液に吸収させる方法や活性炭などの吸着剤に吸着させる方法などが夫々提案されている。

これらの方法のうち、吸着剤を用いる方法は維持が簡単で、ある程度大容量のガスにも適用でき、又除去下限も割合い低いなどの利点を有しているようであるが、ただ吸着剤として吸着能力の高い優れたものを必要とする。しかし乍ら、現在この様な条件に適するものが見当らず、一部に使用されている活性炭もその性能持続期間(ライフ)が短かく工業的な使用に対しては不適当である。

本発明者らは大気中、あるいは各種の製品気体中の水銀を除去するために、上記の優れた特長をもつ吸着法を採用することとし、それに適する優れた吸着剤を得ることを目的として数多

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くの実験研究を行なつた結果、吸着剤として活性炭に、周期律表の1族及び2族bの金属元素の塩化物を含浸せしめたものが各種気体中の水銀の吸着除去に著しく優れた性能をもつことを見出し、種々検討の結果、本発明を完成したものである。

本発明に云う周期律表の1族及び2族bの金属元素の塩化物としては特にリチウム、ナトリウム、カリウム、銅、亜鉛、カドミウムなどの各金属の塩化物である。又、これらの金属元素の塩化物を担持させるための活性炭は、一般の活性炭でよいが、特にヤシガラ炭が適当であり、又その形状は普通粒状が用いられるが、勿論粉末も使用できる。

活性炭中の塩化物の担持量は0.01～40重量%、好ましくは0.1～20%であり、それらは活性炭に吸着され或は附着されている。

本発明の吸着剤である活性炭は種々の方法によつて製造することができるが、例えば上記の如き塩化物の1種又は2種以上の混合物を水溶液又はアルコール類、ケトン類、その他の極性有機溶媒の溶液とし、この溶液を浸漬或は噴霧して活性炭に含浸させる。この場合溶液中の金属元素の塩化物の濃度は活性炭にこれらの化合物が均一に含浸される程度のものであればよく、一般には0.1～40%、好ましくは1～30%が適当である。浸漬せしめる場合、浸漬する時間は均一含浸に必要な時間でよく、又、減圧下で含浸する場合には更に短時間ですむ。更に乾燥温度及びその時間は含浸した化合物が熱分解を起さない程度であればよく、一般には常圧下、50～110℃、3～10時間が適当であるが、減圧状態下では更に低温、短時間の乾燥が可能である。

本発明活性炭は、水銀の吸着剤の有効成分として使用され、一般のガス用吸着剤と同様に、各種の気－固接触による吸着方式の殆んどいずれをも使用することが可能である。例えば、固定床方式、移動床方式、流動床方式、気体輸送方式などの各種の方式により水銀或は水銀ガスを含有する気体と接触せしめることができる。勿論、本発明はこの使用法によつて制限されることはない。

本発明の吸着剤の性能を公知の吸着剤である活性炭と比較してみると表1の通りである。これより本発明の吸着剤は活性炭に比較して著しく高い吸着性能を有していることが認められる。又、Ⅱ族aの塩化物例えば塩化カルシウム、塩化ストロンチウム、塩化バリウム及び塩化亜鉛も吸着性能を有するが塩化亜鉛には及ばない。尚、この場合の試験方法は実施例1に示した或は準じた方法である。

表1

空気中水銀濃度	破過迄の時間	
	本発明の吸着剤 塩化亜鉛担持	活性炭
約10mg／m³	10時間以上	1秒以下
約250 〃	25分	〃

この様な本発明の吸着剤における驚異的な性能向上の機構は必らずしも明らかではないが、この様な効果は活性炭以外の多孔性吸着剤、例えば活性アルミナ、シリカ、ゼオライト、オイルコークス、パーライト、けいそう土などには見られないことであることから、活性炭独特のものとみられ、活性炭表面の水銀吸着のための活性点又はミクロやマクロな細孔が、これらの1族及び2族bの金属元素の塩化物によつて影響され、水銀吸着速度が増加すると共に、吸着量も増加したことによるものと考えられる。

本発明の方法を更に詳細な実施例で示すと以下の様になるが、本発明は以下の実施例により制限されることはない。

実施例1

ヤシガラ活性炭（20～50メッシュ）50部を秤取し、塩化亜鉛の10%溶液200部中に浸漬し、充分に攪拌含浸させ約30分後にとりだし、過剰の溶液を減圧濾過して乾燥器に入れ、105～110℃、10時間乾燥させた。

かくして得られた吸着剤を次にこのものを内径6mmのガラス管中に充填し（充填長さ20mm、充填重量350mg）約30mℓの水銀

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を入れた二口の３００ｍℓの三角フラスコに取付け（他の口は解放）、カラムの他端を水銀蒸気計（ベックマン製、Ｋ－２３型）に連結したのち、先端にエアーポンプをとりつけて流量1.1ℓ／minで作動させた。従つてフラスコ内を通る空気は水銀蒸気（約１０ｍｇ／ｍ³）を伴つてカラムに入り、カラム内の吸着剤層を通る間に水銀は吸着され、水銀が減少したガスは水銀蒸気計を通して外に排出された。ここで蒸気計のフレはガス中の残存水銀量を示し、これをレコーダーで記録することにより、カラムに含まれる吸着剤の水銀吸着挙動を知り得た。この記録図よりその条件における吸着剤の破過時間を読みとり、これを吸着剤の性能の尺度とした。その結果、本実施例の吸着剤は１０時間の通気によつても破過に至らなかつた。そしてカラムを通過してでてきた空気中の水銀濃度は、カラムが破過に達する迄は0.0005ｍｇ／ｍ³（５μｇ／ｍ³）以下であつた。一方活性炭の場合は１秒以下の通気によつて破過に達した。そしてカラムを通過してでてきた空気中の水銀濃度は0.005ｍｇ／ｍ³以上であつた。

実施例２

実施例１に準じヤシガラ活性炭（２０～５０メッシュ）に、表２に示す１族及び２族bの各種の金属元素の塩化物の溶液を含浸させ、乾燥させた。

かくして得られた吸着剤につき実施例１と同様な試験方法により吸着能力の測定を行なつた。又、水銀をいれたフラスコを熱湯で加熱して水銀濃度を約２５０ｍｇ／ｍ³に高め、その吸着能力を測定した。得られた結果は表２の通りである。

表２

担持物	破過迄の時間	
	Ⅰ通気中Hg 10mg/m³の場合	Ⅱ通気中Hg 250mg/m³の場合
塩化リチウム LiCl	１０時間以上	６０
塩化ナトリウム NaCl	〃	４５
塩化カリウム KCl	〃	４０
塩化銅 $CuCl_2$	〃	２０
塩化亜鉛 $ZnCl_2$	〃	２５
塩化カドミウム $CdCl_2$	〃	５０
なし（活性炭のみ）	１秒以内	１秒以内

実施例３

実施例１で作成した吸着剤を径６ｍｍのガラス管に２０ｍｍの長さに充填（約３５０ｍｇ）し、これに水素ガス（Hg約１２ｍｇ／ｍ³を含む）を流速1.1ℓ／minで通し、実施例１と同様な装置によりカラムを通つて出てきた水素ガス中の水銀の分析を行なつた。その結果、１０時間の通気によつても本実施例の吸着剤は破過に至らなかつたが、一方活性炭単独の場合は１秒以内に破過に達した。尚カラムを通過した水素ガス中の水銀の濃度は、本実施例の吸着剤の場合は約0.005ｍｇ／ｍ³（5μｇ／ｍ³）であつたが、活性炭単独の場合は時間が短かく、不安定で、0.005ｍｇ／ｍ³以下とはならなかつた。

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明細書

1 発明の名称

スキーバインデイング

2 特許請求の範囲

対称的に配置された、それぞれ1つの軸を中心として旋回可能である、ほぼアングルレバー状の2つの靴底保持体を有していて、これらの靴底保持体がそれぞれ一方のアームで靴底に接触しかつ他方のアームでばね力の作用下にある、共通の構成部分に作用している形式のスキーバインデイングにおいて、各アングルレバー状の靴底保持体1、2の一方のアームがほぼ垂直な軸4、5を中心として回転可能に支承された複数のローラ6、7で靴底8に接触していて、前記ローラ6、7の内一方のローラ6が靴底側面に近い範囲に、他方のローラ7が靴底端面に近い範囲に設けられていることを特徴とするスキーバインデイング。

3 発明の詳細な説明

本発明は、対称的に配置された、それぞれ1つの軸を中心として旋回可能である、ほぼアングルレバー状の2つの靴底保持体を有していて、これらの靴底保持体がそれぞれ一方のアームで靴底に接触しかつ他方のアームでばね力の作用下にある、共通の構成部分に作用している形式のスキーバインデイングに関する。

軸を中心として旋回可能な2つのアングルレバーを有するスキーバインデイングは公知である。この2つのアングルレバーの一方のアームはばねによつて負荷されており、これによつて他方のアームはスキー靴に向かつて圧着されかつスキー靴が保持されるようになつている。このばあい確実な保持を保証するためにはアームは所定の範囲にわたつてスキー靴に接触しなければならない。側方へ過負荷が生じると、一方のアングルレバーが旋回せしめられかつスキー靴はこのアングルレバーを擦過してアングルレバーから離反しなければならない。勿論このばあいには強い摩擦力が生じ、これによつて相当の摩滅がひき起される。さらにこのばあいに生

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じる摩擦力は表面の状態、例えば表面に水、氷、雪、汚泥が付着していることによつて大きく異なり、したがつてスキー靴の解放が同じ大きさの負荷で行なわれるように調節されていても、スキー靴がきわめて異なる強さの負荷で解放されるようになる。つまりスキー靴の解放は早すぎたりあるいは遅すぎたり、またはまつたく行なわれなくなり、この結果大きな事故が起ることがある。

さらに旋回可能なレバーが端部ならびにレバーの延長に設けられたローラでスキー靴に接触しているスキーバインデイングは既に公知である。この靴底保持体はアングルレバーではなく、ローラの軸に別のレバーが配置されて、このレバーの他方の共通の連接点がばねと結合されている。このばあいにも解放に際してレバーの端部に、外部からの影響を受けて大きく変化する滑り摩擦が生じる。さらにこのばあいの別の欠点は、レバーの基準位置を種々異なる靴底に適合させることができないことである。

本発明の課題は、スキー靴と靴底保持体部分との間の摩擦ができるだけ僅かでありかつ解放過程中にもほぼ一定に保たれるようなスキーバインデイングを提供することである。

この課題は本発明によれば、各アングルレバー状の靴底保持の一方のアームがほぼ垂直な軸を中心として回転可能に支承された複数のローラで靴底に接触していて、前記ローラの内一方のローラが靴底側面に近い範囲に、他方のローラが靴底端面に近い範囲に設けられていることによつて解決された。これによつて解放過程においては転動摩擦だけしか生じなくなる。この転動摩擦は滑り摩擦よりも僅かであるばかりではなく、外部の影響に左右されることも少ない。

さらに本発明の構成によれば、使用状態においては、靴底の端面側の範囲に設けられた両方のローラの軸の間隔は、アングルレバー状の靴底保持体が旋回可能に支承されている軸の間隔よりも小さくなつている。このばあいにはスキー靴によつてローラに押圧力がかけられることによつて、アングルレバー状の靴底保持体を内側に向かつて旋回させようとする回転モーメントが生ぜしめられる。これによつてアングルレバー状の靴底保持体の端部に設けられたローラは横方向にスキー靴底に圧着される。したがつてスキー靴は確実に保持されるだけではなく、自動的に必要な中央位置に保持される。

アングルレバー状の靴底保持体のローラを保持しているアームの間の間隔を選択的に調節するためには、他方のアームはばね力の作用下にある構成部材とバインデイングケーシングに配置された調節ねじとの間に設けられている。これによつて調節ねじを回動させてアングルレバー状の靴底保持体を適当に旋回させることによつて種々異なるスキー靴底にバインデイングを適合させることができるようになる。

次に図面について本発明を説明する：

第1図に略示されているように2つのアングルレバー状の靴底保持体1、2が軸3を中心として旋回可能に支承されている。各アングルレバー状の靴底保持体は一方のアームに2つのほぼ垂直な軸4、5を有している。これらの軸4、5にはローラ6、7が支承されている。スキー靴8は端面側の範囲で、靴底保持体1、2の軸3よりも相互間に僅かな間隔を有する軸5に支承されたローラ7に支えられている。これによつて靴底保持体1、2を軸3を中心として内側に向かつて旋回させようとするモーメントが生じる。したがつてアーム1の端部に設けられたローラ6、7はより強くスキー靴にあてつけられる。

種々異なるスキー靴底の形に適合させるためには靴底保持体1、2の基準位置を変化させることができる。このためには靴底保持体のアーム2はばね9の作用下にあるスライダ10と調節ねじ11との間に突入している。調節ねじ11をバインデイングケーシング12内で回動させることによつて回転方向に応じて靴底保持体は一点鎖線あるいは破線で示された位置に旋回せ

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しめられる。

第2図と第3図に示された構造においては、アングルレバー状の靴底保持体1、2は同様に軸4、5を中心として回転可能に支承されたローラ6、7を保持している。旋回軸3はバインデイングケーシング12に配置されている。このばあいには靴底保持体のアーム2だけが端部に設けられたスライダ10の折曲げ部13に接触しているのではなく、折曲げ部13には靴底押え部材15のアーム14も作用している。靴底押え部材15はバインデイング部材12に設けられた軸16に旋回可能に支承されておりかつ種々異なる靴底厚さに適合させるためにねじ17で高さ調節可能である。

ばね9は一方ではバインデイングケーシング12に、他方ではばね受け18に支えられている。このばね受け18はねじ19によつてスライダ10内で移動可能であるが回転不能に設けられている。ねじ19自体はスライダ10の折曲げ部20の孔内に支えられている。ねじ19を回動させることによつてばね受け18が移動しかつばね9の張力を変化させる。解放力を表示するためにはばね受け18はスライダ10の窓22内に突出する指針21を形成する。

調節ねじ11を回動させることによつてスライダ10もばね9を緊縮しながら一方の方向にあるいはばね9を弛緩しながら他方の方向に移動する。

このばあいにも特にアングルレバー状の靴底保持体の位置に関する表示を行なうために、スライダ10の窓22はケーシング部分24の別の窓23の下に設けられている。したがつて窓23内のマークは靴底保持体位置と調節された解放力とを表示する。

本発明は図示された実施例に限定されるものではなく、種々異なる態様で実施することができる。例えば規格化された靴底を使用するばあいにはアングルレバーの基準位置を変化させる必要はない。さらに必要なばあいには表示を別個に行なうこともできる。

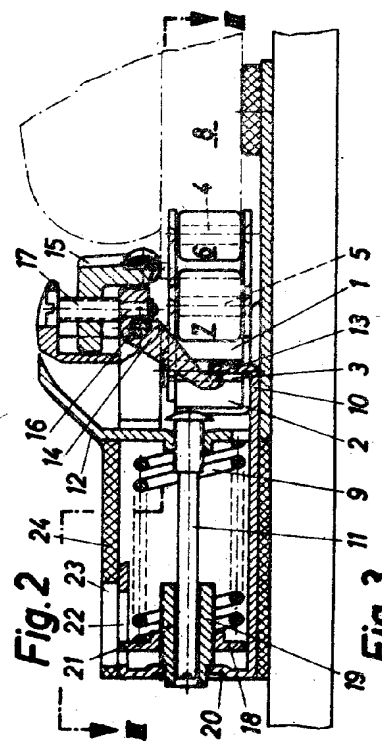
4 図面の簡単な説明

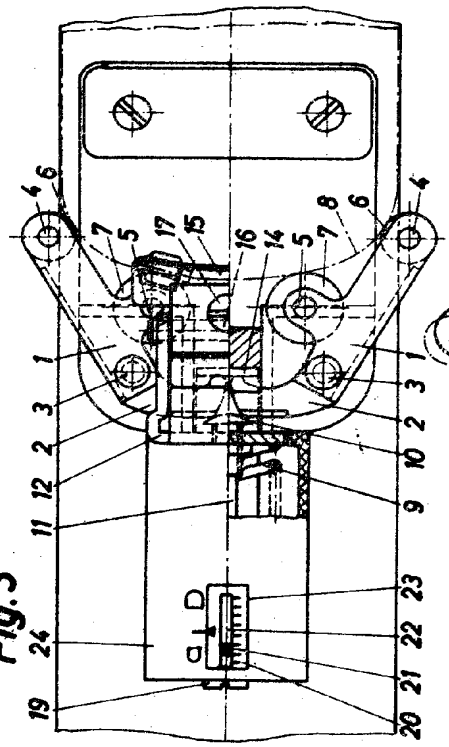
図面は本発明の1実施例を示すものであつて、第1図は本発明のバインデイングの概略図、第2図は本発明のバインデイングの中央縦断面図、第3図の上半分は第2図の平面図、第3図の下半分は第2図のII－II線に沿つた断面図である。

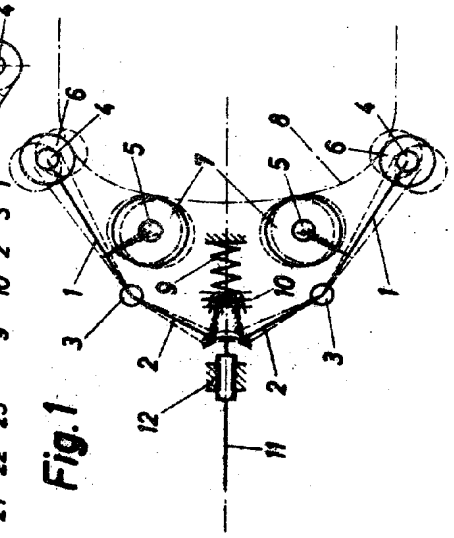
1、2・・靴底保持体、3・・軸、4、5・・軸、6、7・・ローラ、8・・スキー靴、9・・ばね、10・・スライダ、11・・調節ねじ、12・・バインデイングケーシング、13・・折曲げ部、14・・アーム、15・・靴底押え部材、16・・軸、17・・ねじ、18・・ばね受け、19・・ねじ、20・・折曲げ部、21・・指針、22・・窓、23・・窓、24・・ケーシング部分。

代理人 弁護士 ローランド・ゾンデルホフ
（ほか1名）

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Page 121







5. 添附書類の目録

(1)	明細書	1通
(2)	図面	1通
(3)	委任状	1通
(4)	優先権証明書	1通
( )	~~出願審査請求書~~	通

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手続補正書(方式)

適

昭和49年5月13日

特許庁長官殿

1. 事件の表示 昭和48年特許願第 135641 号

2. 発明の名称

スキーバインデイング

3. 補正をする者

事件との関係：特許出願人

名称 ゲルチユ・アクチエンゲゼルシヤフト

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昭和49年4月23日 (発送日)

6. 補正の対象

明細書(第9頁)

7. 補正の内容

別紙の通り
但し、明細書の浄書(内容に変更なし)

特許庁
49.5.14

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4　図面の簡単な説明

図面は本発明の1実施例を示すものであつて、第1図は本発明のバインデイングの概略図、第2図は本発明のバインデイングの中央縦断面図、第3図の上半分は第2図の平面図、第3図の下半分は第2図のⅡ－Ⅱ線に沿つた断面図である。

1、2・・靴底保持体、3・・軸、4、5・・軸、6、7・・ローラ、8・・スキー靴、9・・ばね、10・・スライダ、11・・調節ねじ、12・・バインデイングケーシング、13・・折曲げ部、14・・アーム、15・・靴底押え部材、16・・軸、17・・ねじ、18・・ばね受け、19・・ねじ、20・・折曲げ部、21・・指針、22・・窓、23・・窓、24・・ケーシング部分。

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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

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

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.

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

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

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

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.

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.

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.

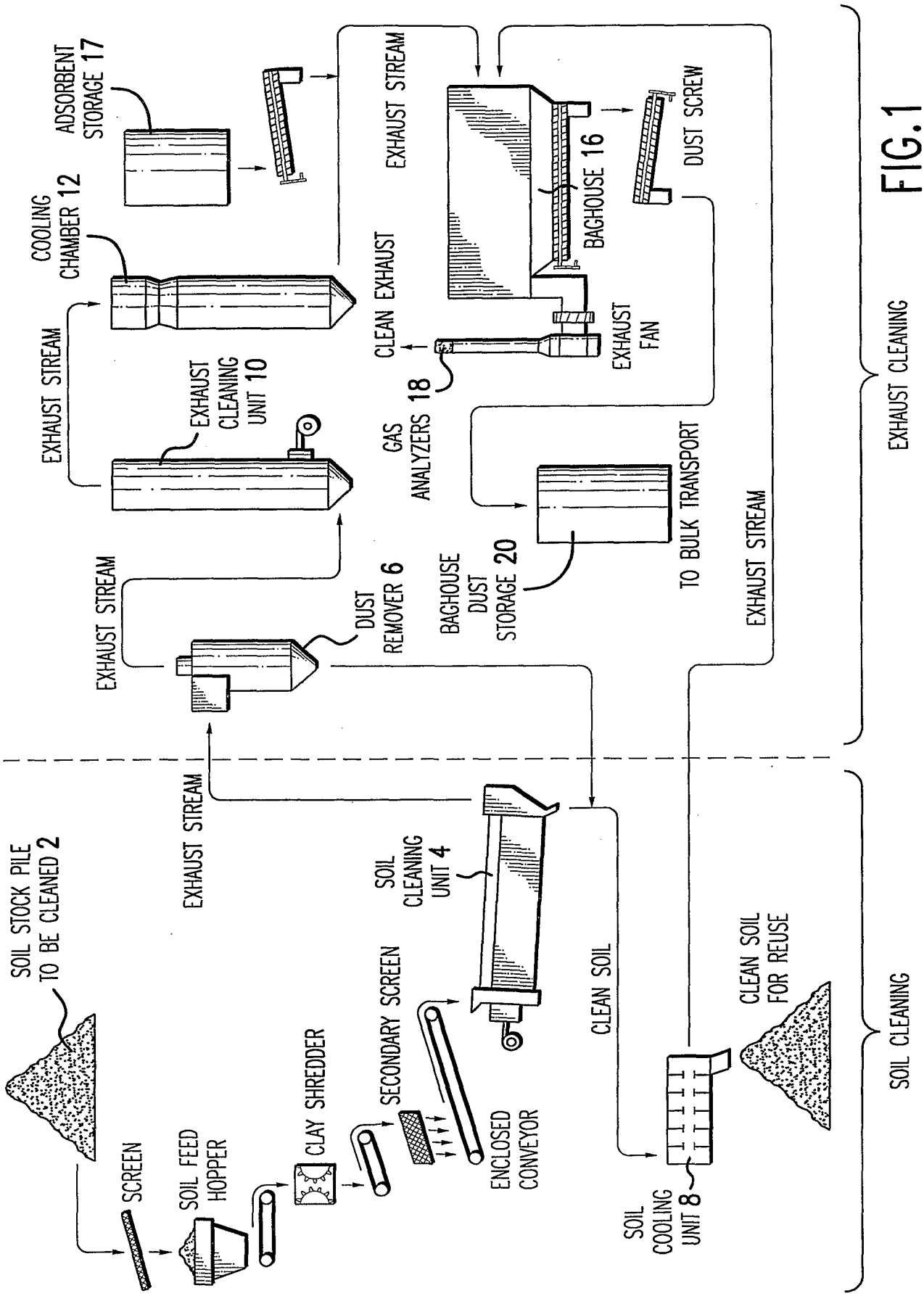

SOIL STOCK PILE TO BE CLEANED 2
SCREEN
SOIL FEED HOPPER
CLAY SHREDDER
SECONDARY SCREEN
ENCLOSED CONVEYOR
SOIL CLEANING UNIT 4
EXHAUST STREAM
SOIL COOLING UNIT 8
CLEAN SOIL
CLEAN SOIL FOR REUSE
SOIL CLEANING
DUST REMOVER 6
EXHAUST STREAM
EXHAUST CLEANING UNIT 10
EXHAUST STREAM
COOLING CHAMBER 12
ADSORBENT STORAGE 17
BAGHOUSE DUST STORAGE 20
TO BULK TRANSPORT
GAS ANALYZERS 18
CLEAN EXHAUST
EXHAUST FAN
BAGHOUSE 16
DUST SCREW
EXHAUST STREAM
EXHAUST STREAM
EXHAUST CLEANING

FIG. 1

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US01/05259

A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) :B01D 53/04
US CL :95/133, 134, 141, 900; 96/154; 423/210
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. : 95/133, 134, 141, 142, 900; 96/108, 132, 135, 154; 423/210

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)

NONE

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 3,193,987 A (MANES et al.) 13 July 1965 (13.07.65).	1-9
A	US 3,194,629 A (DREIBELBIS et al.) 13 July 1965 (13.07.65).	1-9
A	US 3,876,393 A (KASAI et al.) 08 April 1975 (08.04.75).	1-9
A	US 4,094,777 A (SUGIER et al.) 13 June 1978 (13.06.78)	1-9
A	US 4,273,747 A (RASMUSSEN) 16 June 1981 (16.06.81)	1-9
A	US 5,409,522 A (DURHAM et al.) 25 April 1995 (25.04.95).	1-9

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

*	Special categories of cited documents:	"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
"A"	document defining the general state of the art which is not considered to be of particular relevance	"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
"E"	earlier document published on or after the international filing date	"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
"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)	"&"	document member of the same patent family
"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		

Date of the actual completion of the international search 09 APRIL 2001	Date of mailing of the international search report 07 MAY 2001
Name and mailing address of the ISA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. (703) 305-3230	Authorized officer ROBERT H. SPITZER Telephone No. (703) 308-0651

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US01/05259

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5,575,982 A (REISS et al.) 19 November 1996 (19.11.96).	1-9
A	US 5,607,496 A (BROOKS) 04 March 1997 (04.03.97).	1-9
A	US 5,659,110 A (HERDEN et al.) 19 August 1997 (19.08.97).	1-9

Electronic Patent Application Fee Transmittal

Application Number:	12201595
Filing Date:	29-Aug-2008
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Filer:	Dennis Robert Daley/Rebecca Krueger
Attorney Docket Number:	16291.3USD1

Filed as Large Entity

Utility under 35 USC 111(a) Filing Fees

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Pages:				
Claims:				
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
Extension-of-Time:				

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Miscellaneous:				
Submission- Information Disclosure Stmt	1806	1	180	180
		Total in USD ($)		**180**

Electronic Acknowledgement Receipt

EFS ID:	6546335
Application Number:	12201595
International Application Number:	
Confirmation Number:	9735
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Customer Number:	23552
Filer:	Dennis Robert Daley/Rebecca Krueger
Filer Authorized By:	Dennis Robert Daley
Attorney Docket Number:	16291.3USD1
Receipt Date:	01-DEC-2009
Filing Date:	29-AUG-2008
Time Stamp:	12:41:14
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	Credit Card
Payment was successfully received in RAM	$180
RAM confirmation Number	14363
Deposit Account	132725
Authorized User	DALEY,DENNIS R.

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

Charge any Additional Fees required under 37 C.F.R. Section 1.21 (Miscellaneous fees and charges)

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	Information Disclosure Statement (IDS) Filed (SB/08)	IDS.PDF	247120 54cd6ae308308839b85678a0c9704e1aadc396f9	no	5
Warnings:					
Information:					
This is not an USPTO supplied IDS fillable form					
2	NPL Documents	AttachmentA.PDF	6738855 254b7a456945483a347f701d70a1ff81f6617b9e	no	106
Warnings:					
Information:					
3	NPL Documents	AttachmentB.PDF	251213 aa2ca50f1a876c19d2c2d591b288ba659d8a307f	no	7
Warnings:					
Information:					
4	Foreign Reference	CA2150529.PDF	817971 bb4cfc4221f5695f674bf70ab4cdaa8d482979c4	no	25
Warnings:					
Information:					
5	Foreign Reference	DE3426059.PDF	335197 fd166769f1b10bf0d7e10302acdefc867f62b9ca	no	10
Warnings:					
Information:					
6	Foreign Reference	DE10233173.PDF	1203266 68f117489fd44a451a89d066d5b9ab32f5ca33a3	no	20
Warnings:					
Information:					
7	Foreign Reference	EP0208490.PDF	564646 df3f7ba568377bbd31cd6c555bf555aeee0e3d23	no	14
Warnings:					
Information:					
8	Foreign Reference	JP4943197.PDF	194669 e9da5e3f1999c525cdebbeee5b98132dabbe5658	no	3
Warnings:					
Information:					

9	Foreign Reference	JP4953593-W-Abstract.PDF	185302 5d137d10e5ae4869a62220ffc0154b9991afdcc1	no	4
Warnings:					
Information:					
10	Foreign Reference	JP506438.PDF	285430 5af36f4af44be094ca6cf7846bd949d4a550d180	no	5
Warnings:					
Information:					
11	Foreign Reference	WO01_62368.PDF	674867 0c77cc3a5e198f398288eac692228697f5ec4fca	no	14
Warnings:					
Information:					
12	NPL Documents	Dunham-Mercury-Capture.PDF	869729 2d2925312083e964103ba0aecf758a6534b74562	no	6
Warnings:					
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Information:					
13	NPL Documents	Felsvang-Mercury-Reduction.PDF	529923 ccb44a308180a9305f0a5328c87a1250f64c69c9	no	19
Warnings:					
Information:					
14	NPL Documents	Ghorishi-Sorption-of-Mercury.PDF	1592606 14087653a3a6d23613c76ef4a1c7ae3f4fb741e0	no	12
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15	NPL Documents	Lancia-Adsorption-of-Mercury.PDF	1694247 a4bcd819b433fddd596d13fbdc2b9960afd8b3a2	no	14
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Information:					
16	NPL Documents	Olson-The-Reduction-of-Gas.PDF	131348 17676c623ca211e3eba3dac56b6b1c819ba5c560	no	13
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Information:					
17	NPL Documents	Vosteen-Chlor.PDF	1425947 67502e7f0c09e81dfae94ef092baa1368edcd5a3	no	31
Warnings:					
Information:					
18	Fee Worksheet (PTO-875)	fee-info.pdf	30413 f3e5bdae4f630d05323da771caa1b30c55d65b86	no	2
Warnings:					
Information:					
			Total Files Size (in bytes):	17772749	

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

New Applications Under 35 U.S.C. 111

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

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

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

New International Application Filed with the USPTO as a Receiving Office

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

UNITED STATES PATENT AND TRADEMARK OFFICE

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APPLICATION NUMBER	FILING OR 371(C) DATE	FIRST NAMED APPLICANT	ATTY. DOCKET NO./TITLE
12/201,595	08/29/2008	Edwin S. Olson	16291.3USD1

CONFIRMATION NO. 9735

23552
MERCHANT & GOULD PC
P.O. BOX 2903
MINNEAPOLIS, MN 55402-0903

POA ACCEPTANCE LETTER

OC00000038397734

Date Mailed: 10/29/2009

NOTICE OF ACCEPTANCE OF POWER OF ATTORNEY

This is in response to the Power of Attorney filed 10/16/2009.

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

/fstephanos/

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

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant:	OLSON ET AL.	Examiner:	A. ORLANDO
Serial No.:	12/201,595	Group Art Unit:	1797
Filed:	AUGUST 29, 2008	Docket No.:	16291.3USD1
Title:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY		

EFS Web Transmission

REVOCATION AND POWER OF ATTORNEY

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

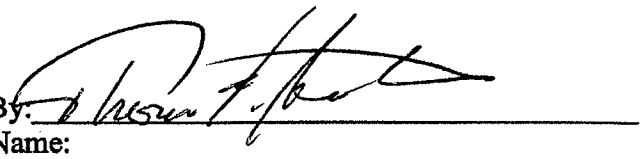
Dear Sir:

Please revoke any existing Powers of Attorney, if any, and appoint the following attorneys and/or patent agents associated with the following customer number to prosecute this application and to transact all business in the U.S. Patent and Trademark Office in connection therewith:

23552
PATENT TRADEMARK OFFICE

Please direct all correspondence in this application to the address associated with the customer number referenced above.

Dated: September 22, 2009

By: 
Name:
Title:

S/N 12/201,595 PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant:	OLSON ET AL.	Examiner:	A. ORLANDO
Serial No.:	12/201,595	Group Art Unit:	1797
Filed:	AUGUST 29, 2008	Docket No.:	16291.3USD1
Title:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY		

CERTIFICATE UNDER 37 C.F.R. § 3.73(b)

Energy & Environmental Research Center Foundation, having a place of business at 15 North 23rd Street, Grand Forks, North Dakota 58202, certifies that it is the assignee of the entire right, title and interest in the patent application identified above by virtue of either:

A. ☒ An assignment from the inventor(s), of the patent application identified above. The assignment was recorded in the Patent and Trademark Office on March 16, 2007, at Reel 019023, Frame(s) 0539, or for which a copy is attached.

B. ☐ A chain of title from the inventor(s) of the patent application identified above to the current assignee as shown below:

1. From: To:

The document was recorded in the Patent and Trademark Office at Reel , Frame(s) , or for which a copy thereof is attached.

2. From: To:

The document was recorded in the Patent and Trademark Office at Reel , Frame(s) , or for which a copy thereof is attached.

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☐ Additional documents in the chain of title are listed on a supplemental sheet.

☐ Copies of assignments or other documents in the chain of title are attached.

The undersigned (whose title is supplied below) is empowered to act on behalf of the assignee.

I hereby declare that all statements made herein of my own knowledge are true, and that all statements made on information and belief are believed to be true; and further, that these statements are made with the knowledge that willful false statements, and the like so made, are punishable by fine or imprisonment, or both, under Section 1001, Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: September 22, 2009 By: _____

Name:

Title:

23552
PATENT TRADEMARK OFFICE

Electronic Patent Application Fee Transmittal

Application Number:	12201595
Filing Date:	29-Aug-2008
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Filer:	Dennis Robert Daley/Sage Kruse
Attorney Docket Number:	2380-00207

Filed as Small Entity

Utility under 35 USC 111(a) Filing Fees

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Pages:				
Claims:				
Miscellaneous-Filing:				
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
Extension-of-Time:				
Extension - 3 months with $0 paid	2253	1	555	555

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Miscellaneous:				
			Total in USD ($)	**555**

Electronic Acknowledgement Receipt

EFS ID:	6278808
Application Number:	12201595
International Application Number:	
Confirmation Number:	9735
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Customer Number:	23505
Filer:	Dennis Robert Daley/Sage Kruse
Filer Authorized By:	Dennis Robert Daley
Attorney Docket Number:	2380-00207
Receipt Date:	16-OCT-2009
Filing Date:	29-AUG-2008
Time Stamp:	16:28:59
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	Credit Card
Payment was successfully received in RAM	$555
RAM confirmation Number	2509
Deposit Account	132725
Authorized User	DALEY,DENNIS R.

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

Charge any Additional Fees required under 37 C.F.R. Section 1.16 (National application filing, search, and examination fees)

Charge any Additional Fees required under 37 C.F.R. Section 1.17 (Patent application and reexamination processing fees)

Charge any Additional Fees required under 37 C.F.R. Section 1.19 (Document supply fees)

Charge any Additional Fees required under 37 C.F.R. Section 1.20 (Post Issuance fees)

Charge any Additional Fees required under 37 C.F.R. Section 1.21 (Miscellaneous fees and charges)

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1		Amendment.pdf	940750 d744ccbbe464f38666b8c4fc7eeaaa850e532647	yes	18
	Multipart Description/PDF files in .zip description				
	Document Description		**Start**	**End**	
	Claims		1	3	
	Applicant Arguments/Remarks Made in an Amendment		4	18	
Warnings:					
Information:					
2	Extension of Time	EOT.pdf	35194 773cb90c40bb2019252d29ffb199bca617553745	no	1
Warnings:					
Information:					
3	Power of Attorney	RevPOA.pdf	99724 1d170b5990507e230c5068b5dac0a740be7765d0	no	3
Warnings:					
Information:					
4	Fee Worksheet (PTO-875)	fee-info.pdf	30339 a21deafa22f540bcc20c22f7a3b412ff86c91ae3	no	2
Warnings:					
Information:					
		Total Files Size (in bytes):	1106007		

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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.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant:	OLSON ET AL.	Examiner:	A. ORLANDO
Serial No.:	12/201,595	Group Art Unit:	1797
Filed:	AUGUST 29, 2008	Docket No.:	16291.3USD1
		Confirmation No.	9735
Title:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY		

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AMENDMENT AND RESPONSE

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

Dear Sir:

This is in response to the Office Action mailed April 16, 2009. Entry of the following amendment and reconsideration of the outstanding rejection are requested.

Amendments to the Claims are reflected in the listing of claims that begins on page 2 of this paper.

Support for Amendment begins on page 4 of this paper.

Remarks begin on page 6 of this paper.

Amendments to the Claims:

This listing of claims will replace all prior versions and listings of claims in the application.

Listing of Claims:

1. (Currently amended) 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, the promoted carbon sorbent comprising a reaction product of the promoter and a carbene site on the base activated carbon to form a carbocation accompanied by a halide anion wherein the promoted carbon sorbent is effective for the removal of mercury from a gas stream ~~located downstream of a boiler, said removal of mercury yielding a cleaned gas stream, and wherein the composition of the promoted carbon sorbent is controlled by the mercury content of the cleaned gas stream~~.

2. (Original) 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. (Currently amended) The promoted carbon sorbent of claim 2 wherein the sorbent comprises an activated carbon matrix in a fixed- or a moving-bed structure consisting of moving fiber screens and monoliths.

4-6. (Cancelled).

7. (Currently Amended) The promoted carbon sorbent of claim 2 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.

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Page 154

8-9. (Cancelled)

10. (Currently amended) The promoted carbon sorbent of claim 2 further comprising a ~~an optional~~ secondary component comprising a halogen, ~~or~~ a hydrohalide, a Group V halide, a Group VI halide, or a combination thereof ~~such that the reactivity and mercury capacity of the sorbent are enhanced~~.

11-13. (Cancelled)

14. (Currently amended) The promoted carbon sorbent of claim ~~13~~1 wherein the promoter is substantially in gas phase ~~vapor form~~ when combined with the base activated carbon.

15. (Currently Amended) The promoted carbon sorbent of claim ~~13~~10 wherein the ~~promoter is combined with an organic solvent prior to reaction with the base activated carbon~~secondary component is substantially in gas phase when combined with the promoted carbon sorbent.

16. (Currently amended) The promoted carbon sorbent of claim ~~13~~10 wherein the promoter and ~~optional~~ secondary component are combined with the base activated carbon substantially simultaneously.

17-33. (Cancelled)

34. (New) The promoted carbon sorbent of claim 1 wherein the promoted carbon sorbent oxidizes and captures mercury from a gas stream.

35. (New) The promoted carbon sorbent of claim 1 wherein the promoted carbon sorbent is effective for the removal of at least one of boron, tin, arsenic, gallium, antimony, lead, bismuth, cadmium, silver, copper, and zinc.

Support for Amendment

Claim 1 is amended to characterize the promoted carbon sorbent as "comprising a reaction product of the promoter and a carbene site on the base activated carbon to form a carbocation accompanied by a halide anion. This amendment is supported by the specification at paragraph 10 which describes the reaction product of base activated carbon and promoter, and that the reaction product is effected for the removal of mercury from a gas stream. In addition, paragraph 82 of the specification characterizes the reaction as accruing on the carbene site of the base activated carbon to form a carbocation. In addition, claim 1 is amended to characterize the sorbent as a "promoted carbon sorbent" to more clearly provide antecedent basis. In addition, claim 1 is amended to remove the reference to where the removal of mercury from a gas stream occurs because the claim is directed at a promoted carbon sorbent rather than a method of removing mercury from a gas stream.

Claim 3 is amended to include the features of cancelled claim 4.

Claims 10 and 16 are amended to remove the word "optional."

Claim 7 is amended to remove SO_2 from the lists.

Claim 10 is amended by removing the phrase "such that the reactivity and mercury capacity of the sorbent are enhanced" and to include in the list of secondary component "a Group V halide, a Group VI halide, or a combination thereof." This amendment is supported by the specification at paragraph 23, page 5.

Claim 14 is amended to change the phrase "vapor form" to "gas phase." This amendment is supported by the specification at, for example, paragraph 50, page 9.

Claim 15 is amended to characterize the secondary component as substantially in gas phase when combined with the promoted carbon sorbent. This amendment is supported by the specification at, for example, paragraph 50, page 9.

New claim 34 characterizes the promoted carbon sorbent as a promoted carbon sorbent that oxidizes and captures mercury in a gas stream. New claim 35 characterizes the promoted carbon sorbent as a promoted carbon sorbent that is effective for the removal of at least one of boron, tin, arsenic, gallium, antimony, lead, bismuth, cadmium, silver, copper, and zinc. Support for new claims 34 and 35 can be found in the specification at paragraph 81, pages 17-18.

Claims 14-15 are amended to change dependency.

No new matter is introduced by this amendment, and entry thereof is requested. Upon

entry claims 1-3, 7, 10, 14-16, 34, and 35 are active in this application.

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Page 157

REMARKS

Applicants acknowledge receipt of the nonfinal *Office Action* mailed April 16, 2009, in which the Examiner: (1) objected to claim 21 as being of improper dependant form for failing to limit the subject matter of a previous claim; (2) rejected claims 4 and 24 as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention; (3) rejected claims 1, 17, 18, and 20–22 under 35 U.S.C. § 102(a) over *Srinivasachar* et al. (U.S. Patent No. 6,848,374) (*Srinivasachar*); (4) rejected claims 25–27, 29–31, and 33 under 35 U.S.C. § 103(a) over *Srinivasachar*; (5) rejected claim 2 under 35 U.S.C. § 103(a) over *Srinivasachar* and *Reiss et al.* (U.S. Patent No. 5,575,982) *(Reiss)*; (6) rejected claims 3-6 under 35 U.S.C. § 103(a) over *Srinivasachar, Reiss,* and *Moller et al.* (U.S. Patent No. 4,889,698) (*Moller*); (7) rejected claims 7 and 8 under 35 U.S.C. § 103(a) over *Srinivasachar, Reiss*, and *Broderick* (U.S. Patent No. 6,942,840) *Broderick*; (8) rejected claim 9 under 35 U.S.C. § 103(a) over *Srinivasachar, Reiss*, *Broderick,* and *Moller*; (9) rejected claim 10-16 under 35 U.S.C. § 103(a) over *Srinivasachar, Reiss*, and *Moller*; (10) rejected claim 19 under 35 U.S.C. § 103(a) over *Srinivasachar* and *Reiss;* (11) rejected claims 23 and 24 under 35 U.S.C. § 103(a) over *Srinivasachar* and *Moller et al.* (U.S. Patent No. 4,889,698) (*Moller*); (12) rejected claim 28 under 35 U.S.C. § 103(a) over *Srinivasachar* and *Moller;* (13) rejected claim 32 under 35 U.S.C. § 103(a) over *Srinivasachar* and *Moller.*

Applicants respond as follows.

1. Objection to claim 21 under 37 CFR 1.75(c)

In the Office Action claim 21 is objected to under 37 CFR 1.75(c). Claim 21 has been canceled. Withdrawal of the rejection is requested.

2. Rejected claims 4 and 24 under 35 U.S.C. §112, second paragraph

In the Office Action, claims 4 and 24 are rejected under 35 USC §112, second paragraph. Claims 4 and 24 have been canceled. Withdrawal of the rejection is requested.

3. Rejected claims 1, 17, 18, and 20–22 under 35 U.S.C. §102(a) over Srinivasachar

The Office Action includes a rejection of claims 1, 17, 18, and 20–22 under 35 U.S.C. §102(b) over *Srinivasachar*. This rejection is traversed.

The present invention is directed at a promoted carbon sorbent. The promoted carbon sorbent comprises a base activated carbon reacted with a promoter selected from the group consisting of halides, halogens, and combinations thereof. The promoted carbon sorbent comprises a reaction product of the promoter and a carbene site on the base activated carbon to form a carbocation accompanied by a halide anion. The promoted carbon sorbent is effective for the removal of mercury from a gas stream.

Srinivasachar fails to describe a promoted carbon sorbent comprising a reaction product of a promoter selected from the group consisting of halides, halogens, and combinations thereof, and a carbene site on a base activated carbon to form a carbocation accompanied by a halide anion according to the presently claimed invention.

Srinivasachar is directed at the production of a carbonaceous sorbent for the capture of elemental mercury. In addition, *Srinivasachar* is directed at the "treatment of the carbonaceous sorbent to enhance the capture of elemental mercury." See *Srinivasachar* at column 2, line 10-18. The treatment of the carbonaceous sorbent involves "condensing/adsorbing" a halogen 58 onto a sorbent 28. See *Srinivasachar* at column 6, lines 13-22, and figure 1. Because *Srinivasachar* describes "condensing/adsorbing" the halogen onto the sorbent, there is clearly no intention by *Srinivasachar* of reacting the halogen and the sorbent. Furthermore, *Srinivasachar* fails to describe conditions that provide for reacting a promoter and a carbene site on a base activated carbon to form a carbocation accompanied by a halide anion according to the presently claimed invention.

Srinivasachar states:

> "the sorbent particles 28 are sent along a path 38 from the silo 30 to oxidizer unit 40, where the particles are contacted by an oxidizing agent 42 (e.g., ozone, hydrogen peroxide, hot air, concentrated nitric acid), making the outer surfaces of the particles hydrophilic. The treated sorbent may then be fed along a path 44 to the separation device 34 or sent along a path 46 to a blender 48 where a solution 50 is sprayed 52 on the sorbent particles by a sprayer 54 to deposit a halogen on the surface of the sorbent particles 28."

See *Srinivasachar* at column 5, line 65 through column 6, line 13. Clearly, *Srinivasachar* describes a process where the sorbent particles are treated to render the outer surfaces of the particles hydrophilic so that the spray of halogen adsorbs onto the hydrophilic surface of the

particles. The solution 50 described by *Srinivasachar* at column 6, lines 7-10, can include halogen acids dissolved in water. It is submitted that this type of aqueous solution would condense and adsorb onto the particles according to *Srinivasachar*.

No sufficient reason or explanation has been provided in the outstanding Office Action that would suggest that the disclosure by *Srinivasachar* would result in a reaction of the halogen with the sorbent particles. Furthermore, there are several reasons why the sorbent particles and halogen acids dissolved in water described by *Srinivasachar* would not react. Those reasons are explained below.

Srinivasachar clearly describes depositing a halogen on the surface of the sorbent particles, and "condensing/adsorbing" the halogen onto the sorbent. Nowhere does *Srinivasachar* mention a reaction between the halogen and the sorbent, and nowhere does *Srinivasachar* recognize an advantage of reacting a promoter and a carbene site on a base activated carbon to form a carbocation accompanied by a halide anion to provide a promoted carbon sorbent according to the presently claimed invention.

> The outstanding Office Action makes the following statement about *Srinivasachar*: "The reference does not explicitly state the carbon sorbent and the promoter reacting. It would have been obvious to one of ordinary skill in the art at the time the invention was made that contacting the activated carbon with the promoter would inherently produce the same results as that in the present invention (e.g. cause a reaction between the two)."

See the outstanding Office Action at page 3, paragraph 5. It is understood that the Examiner's position is that a reaction between the halogen and the sorbent is inherent in the disclosure by *Srinivasachar* although the Examiner does not explain why the conditions of *Srinivasachar* lend itself to the existence of a reaction between the halogen and the sorbent. It is submitted that there is no reason to believe that the reaction is inherent, and there are several reasons why the reaction is not inherent. *Srinivasachar* teaches that the sorbent particles are treated with an oxidizing agent to increase the hydrophilicity of the outer surface of the sorbent particles. Oxidizing agents would add oxygen to the exterior surface of the sorbent particles to enhance the "condensing/adsorbing" of halogen to the sorbent particles. The conditions described by *Srinivasachar* simply provide for depositing a halogen on the surface of the sorbent particles contacted by an oxidizing agent. According to *Srinivasachar*:

"a solution 50 is sprayed 52 on the sorbent particles by a sprayer 54 to deposit a halogen on the surface of the sorbent particles 28. The solution 50 is chosen from potassium iodide, iodine dissolved in potassium iodide, alkali halides (e.g., NaCl), and halide salts (e.g., $CaCl_2$), or halogen acids (e.g., HCl, HI, HBr, HF) dissolved in water."

See *Srinivasachar* at column 6, lines 4-12. Clearly, *Srinivasachar* does not take steps to provide a reaction product resulting from a reaction of a promoter and a carbene site on a base activated carbon to form a carbocation accompanied by a halide anion according to the presently claimed invention. In fact, *Srinivasachar* describe spraying the solution of halogen acid dissolved in water on to carbon black, and carbon black is not a material that contains a carbene site for reaction. The Examiner's attention is directed to *Srinivasachar* at column 6, lines 23-55. At this location of *Srinivasachar*, carbon black and activated carbon are both mentioned. In the case of carbon black, mercury removal performance improved from 20% to 100%. For activated carbon, the mercury removal performance improved from 75% to 90%. Clearly, this shows that higher performance resulted from the use of carbon black as the sorbent as opposed to activated carbon as the sorbent. This means that *Srinivasachar* has not achieved the benefit of reacting the halogen to the carbene site on the activated carbon. Had *Srinivasachar* achieved a reaction between the halogen and the carbene site on the activated carbon, *Srinivasachar* would have seen greater improvement. The Examiner's attention is additionally directed to *Srinivasachar* at column 9, lines 33-52. This portion of *Srinivasachar* clearly reflects a preference for utilizing carbon black rather than activated carbon. One skilled in the art would clearly understand that this means that *Srinivasachar* do not recognize an advantage of reacting halogens with a carbene site on base activated carbon to perform a carbocation accompanied by a halide anion where the promoted carbon sorbent is effective for the removal of mercury from a gas stream according to the presently claimed invention.

Based on the discussions of *Srinivasachar* relating to the use of carbon black and activated carbon as the sorbent, one can conclude that *Srinivasachar* does not achieve a reaction between halogen and the carbene site on activated carbon. Because *Srinivasachar* does not recognize an improvement when using activated carbon compared with carbon black, it is clear that no reaction is achieved by *Srinivasachar* between the halogen and the carbene sites on the activated carbon. One skilled in the art would appreciate that the reason for this relates to the

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treatment of the sorbent particles to make the outer surfaces of the sorbent particles hydrophilic, and the deposition of halogen acid in water on the sorbent particles. It is submitted that this process described by *Srinivasachar* makes it difficult for there to be a reaction between the halogen and a carbene site on activated carbon.

The above-identified patent application provides at least a couple of techniques for controlling the conditions of contact between the promoter and the base activated carbon to provide a reaction product. The Examiner's attention is directed to the specification at paragraph 50 (page 9) which provides that the halogen or halide promoter that is added to, and reacts with, the base activated carbon can be molecular halogen in vapor or gaseous form, molecular halogen in organic solvent, 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). The above-identified patent application at paragraph 51 (page 9) describes an exemplary organic solvent. Furthermore, the above-identified patent application at paragraph 60 (page 12) refers to the "in-flight" preparation of a promoted carbon sorbent where the promoter in vapor or gaseous form is combined with the base activated carbon to provide a reaction. The "in-flight" process is further described in the above-identified patent application at paragraph 75 (page 16). In addition, the above-identified patent application refers to the reaction of the promoter in liquid form or in a solvent (and then with solvent removal) with the base activated carbon to provide a reaction at paragraph 60 (page 12).

The above-identified patent application describes how to obtain a reaction product of a promoter and base activated carbon. In contrast, *Srinivasachar* fails to indicate any interest in obtaining a reaction between a promoter and a sorbent, and fails to describe conditions that would achieve a reaction between a promoter and a sorbent to provide a promoted carbon sorbent according to the presently claimed invention. Accordingly, a promoted carbon sorbent according to the presently claimed invention would not inherently result from *Srinivasachar*.

In view of the above comments, the claimed invention is not anticipated by *Srinivasachar*. Accordingly, withdrawal of the rejection over *Srinivasachar* is requested.

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4. Rejected claims 25–27, 29–31, and 33 under 35 U.S.C. § 103(a) Srinivasachar.

The Office Action includes a rejection of claims 25-27, 29-31, and 33 under 35 U.S.C. §103(a) over *Srinivasachar*. This rejection is traversed.

It is pointed out that claims 25-27, 29-31, and 33 have been cancelled. Nevertheless, the claims upon which these claims depend would not have been obvious over *Srinivasachar*.

Any rejection under 35 U.S.C. § 103 must clearly and explicitly articulate the reason(s) why the claimed invention would have been obvious. MPEP § 2142 (2007). The framework for determining obviousness under 35 U.S.C. § 103 requires (1) determination of the scope and content of the prior art; (2) assessment of the differences between the claimed invention and the prior art; and (3) assessment of the level of ordinary skill in the pertinent art. MPEP § 2141 (2007) (*Graham v. John Deere Co.*, 383 U.S. 1, 148 USPQ 459 [1966]). According to MPEP § 2141, ascertaining the differences between the claimed invention and the prior art requires interpreting the claim language. For instance, to establish obviousness, each of the claim limitations must be taught or suggested by the prior art. See *CFMT, Inc. v. YieldUp Int'l Corp.*, 349 F.3d 1333, 1342 (Fed. Cir. 2003) (citing *In re Royka*, 490 F.2d 981, 985 [CCPA 1974]).

As discussed above, *Srinivasachar* fails to describe a promoted carbon sorbent comprising a reaction product of a promoter selected from the group consisting of halides, halogens, and combinations thereof, and a carbene site on a base activated carbon to form a carbocation accompanied by a halide anion according to the presently claimed invention. Instead, *Srinivasachar* describes contacting sorbent particles with an oxidizing agent to make the outer surface of the particles hydrophilic, and then condensing/adsorbing a halogen onto the sorbent particles. See *Srinivasachar* at column 5, line 62 through column 6, line 22. *Srinivasachar* fails to describe a reaction between the halogen and the sorbent particles, and fails to describe conditions that would result in a reaction between the halogen and the sorbent particles. *Srinivasachar* fails to appreciate a performance advantage from using activated carbon (with a carbene site) over carbon black (not containing a carbene site). This shows that *Srinivasachar* does not achieve a reaction between the aqueous halogen and the sorbent. In contrast, the above-identified patent application explains how to obtain a reaction product between a promoter and a base activated carbon to form a promoted carbon according to the presently claimed invention.

One skilled in the art would not have received a suggestion from *Srinivasachar* to alter

the conditions of "condensing/adsorbing" a halogen onto sorbent particles to achieve a reaction and a promoted carbon sorbent according to the presently claimed invention.

In view of the above comments, the claimed invention would not have been obvious from *Srinivasachar*, and withdrawal of this rejection is requested.

5. Rejected claim 2 under 35 U.S.C. § 103(a) over Srinivasachar and Reiss

The Office Action includes a rejection of claim 2 under 35 U.S.C. § 103(a) over *Srinivasachar* and *Reiss*. This rejection is traversed.

As discussed above, *Srinivasachar* fails to describe a promoted carbon sorbent comprising a reaction product of a promoter selected from the group consisting of halides, halogens, and combinations thereof, and a carbene site on a base activated carbon to form a carbocation accompanied by a halide anion according to the presently claimed invention. Instead, *Srinivasachar* describes contacting sorbent particles with an oxidizing agent to make the outer surface of the particles hydrophilic, and then condensing/adsorbing a halogen onto the sorbent particles. See *Srinivasachar* at column 5, line 62 through column 6, line 22. *Srinivasachar* fails to describe a reaction between the halogen and the sorbent particles, and fails to describe conditions that would result in a reaction between the halogen and the sorbent particles. *Srinivasachar* fails to appreciate a performance advantage from using activated carbon (with a carbene site) over carbon black (not containing a carbene site). This shows that *Srinivasachar* does not achieve a reaction between the aqueous halogen and the sorbent. In contrast, the above-identified patent application explains how to obtain a reaction product between a promoter and a base activated carbon to form a promoted carbon according to the presently claimed invention.

One skilled in the art would not have received a suggestion from *Srinivasachar* to alter the conditions of "condensing/adsorbing" a halogen onto sorbent particles, to achieve a reaction and a promoted carbon sorbent according to the presently claimed invention.

Reiss is relied upon in the outstanding Office Action for the disclosure of activated carbon having an average particle size greater than 40 μm. See the outstanding Office Action at page 9, and *Reiss* at column 4, lines 59-65. It is submitted that *Reiss*, however, fails to suggest forming a promoted carbon sorbent according to the presently claimed invention. Furthermore, one having ordinary skill in the art would not have received a suggestion from *Reiss* to modify

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the conditions described by *Srinivasachar* to achieve a reaction product between the halogen and the sorbent particles described by *Srinivasachar*.

In view of the above comments, the claimed invention would not have been obvious from *Srinivasachar* and *Reiss*, and withdrawal of this rejection is requested.

6. Rejected claims 3-6 under 35 U.S.C. § 103(a) over Srinivasachar, Reiss, and Moller

The Office Action includes a rejection of claims 3-6 under 35 U.S.C. § 103(a) over *Srinivasachar*, *Reiss,* and *Moller*. This rejection is traversed.

As discussed above, *Srinivasachar* fails to describe a promoted carbon sorbent comprising a reaction product of a promoter selected from the group consisting of halides, halogens, and combinations thereof, and a carbene site on a base activated carbon to form a carbocation accompanied by a halide anion according to the presently claimed invention. Instead, *Srinivasachar* describes contacting sorbent particles with an oxidizing agent to make the outer surface of the particles hydrophilic, and then condensing/adsorbing a halogen onto the sorbent particles. See *Srinivasachar* at column 5, line 62 through column 6, line 22. *Srinivasachar* fails to describe a reaction between the halogen and the sorbent particles, and fails to describe conditions that would result in a reaction between the halogen and the sorbent particles. *Srinivasachar* fails to appreciate a performance advantage from using activated carbon (with a carbene site) over carbon black (not containing a carbene site). This shows that *Srinivasachar* does not achieve a reaction between the aqueous halogen and the sorbent. In contrast, the above-identified patent application explains how to obtain a reaction product between a promoter and a base activated carbon to form a promoted carbon according to the presently claimed invention.

One skilled in the art would not have received a suggestion from *Srinivasachar* to alter the conditions of "condensing/adsorbing" a halogen onto sorbent particles, to achieve a reaction and a promoted carbon sorbent according to the presently claimed invention.

Reiss is relied upon in the outstanding Office Action for the disclosure of activated carbon having an average particle size greater than 40 μm. See the outstanding Office Action at page 9, and *Reiss* at column 4, lines 59-65. It is submitted that *Reiss*, however, fails to suggest forming a promoted carbon sorbent according to the presently claimed invention. Furthermore, one having ordinary skill in the art would not have received a suggestion from *Reiss* to modify

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the conditions described by *Srinivasachar* to achieve a reaction product between the halogen and the sorbent particles described by *Srinivasachar*.

Moller is relied upon for the disclosure of activated carbon in a fixed bed. See the outstanding Office Action at page 10, and *Moller* at column 4, lines 39-45. *Moller* is additionally relied upon for the disclosure of alkaline components. See the outstanding Office Action at page 11, and *Moller* at column 7, lines 43-56. This disclosure by *Moller*, however, is not a suggestion to modify *Srinivasachar* to alter the condition of "condensing/adsorbing" a halogen onto sorbent particles, to achieve a reaction and a promoted carbon sorbent according to the presently claimed invention.

In view of the above comments, the claimed invention would have been obvious over *Srinivasachar, Reiss,* and *Moller,* and withdrawal of the rejection is requested.

7. Rejected claims 7 and 8 under 35 U.S.C. § 103(a) over Srinivasachar, Reiss, and Broderick

The Office Action includes a rejection of claims 7 and 8 under 35 U.S.C. § 103(a) over *Srinivasachar*, *Reiss,* and *Broderick*. This rejection is traversed.

As discussed above, *Srinivasachar* fails to describe a promoted carbon sorbent comprising a reaction product of a promoter selected from the group consisting of halides, halogens, and combinations thereof, and a carbene site on a base activated carbon to form a carbocation accompanied by a halide anion according to the presently claimed invention. Instead, *Srinivasachar* describes contacting sorbent particles with an oxidizing agent to make the outer surface of the particles hydrophilic, and then condensing/adsorbing a halogen onto the sorbent particles. See *Srinivasachar* at column 5, line 62 through column 6, line 22. *Srinivasachar* fails to describe a reaction between the halogen and the sorbent particles, and fails to describe conditions that would result in a reaction between the halogen and the sorbent particles. *Srinivasachar* fails to appreciate a performance advantage from using activated carbon (with a carbene site) over carbon black (not containing a carbene site). This shows that *Srinivasachar* does not achieve a reaction between the aqueous halogen and the sorbent. In contrast, the above-identified patent application explains how to obtain a reaction product between a promoter and a base activated carbon to form a promoted carbon according to the presently claimed invention.

One skilled in the art would not have received a suggestion from *Srinivasachar* to alter

the conditions of "condensing/adsorbing" a halogen onto sorbent particles, to achieve a reaction and a promoted carbon sorbent according to the presently claimed invention.

Reiss is relied upon in the outstanding Office Action for the disclosure of activated carbon having an average particle size greater than 40 μm. See the outstanding Office Action at page 9, and *Reiss* at column 4, lines 59-65. It is submitted that *Reiss*, however, fails to suggest forming a promoted carbon sorbent according to the presently claimed invention. Furthermore, one having ordinary skill in the art would not have received a suggestion from *Reiss* to modify the conditions described by *Srinivasachar* to achieve a reaction product between the halogen and the sorbent particles described by *Srinivasachar*.

Broderick is relied upon for the disclosure of a mercury-stabilizing reagent. See the outstanding Office action at page 12. *Broderick* describes several reagents for reacting with vapor phase mercury at column 4, lines 55-65. It is submitted that the disclosure by *Broderick* would not have led one having ordinary skill in the art to modify *Srinivasachar* to alter the conditions of "condensing/adsorbing" a halogen onto sorbent particles, to achieve a reaction and a promoted carbon sorbent according to the presently claimed invention.

In view of the above comments, the claimed invention would have been obvious over *Srinivasachar, Reiss,* and *Broderick,* and withdrawal of the rejection is requested.

8. Rejected claim 9 under 35 U.S.C. § 103(a) over Srinivasachar, Reiss, Broderick, and Moller

The Office Action includes a rejection of claim 9 under 35 U.S.C. § 103(a) over *Srinivasachar, Reiss, Broderick,* and *Moller*. In view of the cancellation of claim 9, it is submitted that this rejection has been rendered moot. Accordingly, withdrawal of this rejection is requested.

9. Rejected claims 10-16 under 35 U.S.C. § 103(a) over Srinivasachar, Reiss, and Moller

The Office Action includes a rejection of claims 10-16 under 35 U.S.C. § 103(a) over *Srinivasachar, Reiss,* and *Moller*. This rejection is traversed.

As discussed above, *Srinivasachar* fails to describe a promoted carbon sorbent comprising a reaction product of a promoter selected from the group consisting of halides, halogens, and combinations thereof, and a carbene site on a base activated carbon to form a carbocation accompanied by a halide anion according to the presently claimed invention.

Instead, *Srinivasachar* describes contacting sorbent particles with an oxidizing agent to make the outer surface of the particles hydrophilic, and then condensing/adsorbing a halogen onto the sorbent particles. See *Srinivasachar* at column 5, line 62 through column 6, line 22. *Srinivasachar* fails to describe a reaction between the halogen and the sorbent particles, and fails to describe conditions that would result in a reaction between the halogen and the sorbent particles. *Srinivasachar* fails to appreciate a performance advantage from using activated carbon (with a carbene site) over carbon black (not containing a carbene site). This shows that *Srinivasachar* does not achieve a reaction between the aqueous halogen and the sorbent. In contrast, the above-identified patent application explains how to obtain a reaction product between a promoter and a base activated carbon to form a promoted carbon according to the presently claimed invention.

One skilled in the art would not have received a suggestion from *Srinivasachar* to alter the conditions of "condensing/adsorbing" a halogen onto sorbent particles, to achieve a reaction and a promoted carbon sorbent according to the presently claimed invention.

Reiss is relied upon in the outstanding Office Action for the disclosure of activated carbon having an average particle size greater than 40 μm. See the outstanding Office Action at page 9, and *Reiss* at column 4, lines 59-65. It is submitted that *Reiss*, however, fails to suggest forming a promoted carbon sorbent according to the presently claimed invention. Furthermore, one having ordinary skill in the art would not have received a suggestion from *Reiss* to modify the conditions described by *Srinivasachar* to achieve a reaction product between the halogen and the sorbent particles described by *Srinivasachar*.

Moller is relied upon for the disclosure of activated carbon in a fixed bed. See the outstanding Office Action at page 10, and *Moller* at column 4, lines 39-45. This disclosure by *Moller*, however, is not a suggestion to modify *Srinivasachar* to alter the condition of "condensing/adsorbing" a halogen onto sorbent particles, to achieve a reaction and a promoted carbon sorbent according to the presently claimed invention.

In view of the above comments, the claimed invention would have been obvious over *Srinivasachar, Reiss,* and *Moller,* and withdrawal of the rejection is requested.

10. Rejected claim 19 under 35 U.S.C. § 103(a) over Srinivasachar and Reiss

The Office Action includes a rejection of claim 19 under 35 U.S.C. § 103(a) over *Srinivasachar* and *Reiss*. In view of the cancellation of claim 19, this rejection has been rendered moot. Accordingly, withdrawal of this rejection is requested.

11. Rejected claims 23 and 24 under 35 U.S.C. § 103(a) over Srinivasachar and Moller

The Office Action includes a rejection of claims 23 and 24 under 35 U.S.C. § 103(a) over *Srinivasachar* and *Moller*. In view of the cancellation of claims 23 and 24, this rejection has been rendered moot. Accordingly, withdrawal of this rejection is requested.

12. Rejected claim 28 under 35 U.S.C. § 103(a) over Srinivasachar and Moller

The Office Action includes a rejection of claim 28 under 35 U.S.C. § 103(a) over *Srinivasachar* and *Moller*. In view of the cancellation of claim 28, this rejection has been rendered moot. Accordingly, withdrawal of this rejection is requested.

13. Rejected claim 32 under 35 U.S.C. § 103(a) over Srinivasachar and Moller

The Office Action includes a rejection of claim 32 under 35 U.S.C. § 103(a) over *Srinivasachar* and *Moller*. In view of the cancellation of claim 32, this rejection has been rendered moot. Accordingly, withdrawal of this rejection is requested.

Conclusion

Applicants respectfully request reconsideration, allowance of the pending claims, and a timely Notice of Allowance be issued in this case. In addition, Applicants believe that this is a full and complete response to each rejection, objection, and requirement. If any item has been overlooked, Applicants respectfully request the opportunity to supplement this response. If the Examiner feels that a telephone conference would expedite the resolution of this case, the Examiner is respectfully requested to contact the undersigned.

In the course of the foregoing discussions, Applicants may have, at times, referred to claim limitations in shorthand fashion or may have focused on a particular claim element. This discussion should not be interpreted to mean that the other limitations can be ignored or dismissed. The claims must be viewed as a whole, and each limitation of the claims must be

considered when determining the patentability of the claims. Moreover, it should be understood that there may be other distinctions between the claims and the prior art that have yet to be raised, but which may be raised in the future.

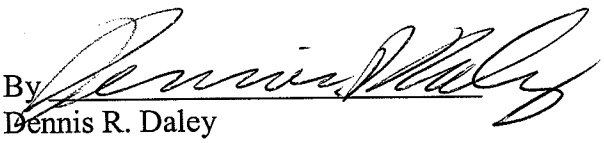
Should any fees have been inadvertently omitted, or if any additional fees are required, or if any fees have been overpaid, please appropriately charge or credit those fees to Deposit Account No. 13-2725, and consider this paper a petition for any necessary extension of time.

Respectfully submitted,

23552
PATENT TRADEMARK OFFICE

MERCHANT & GOULD P.C.
P.O. Box 2903
Minneapolis, Minnesota 55402-0903
(612) 332-5300

Dated: October 16, 2009

By 
Dennis R. Daley
Reg. No. 34,994
DRD/TPJ/cjc/mls

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant:	OLSON ET AL.	Examiner:	A. ORLANDO
Serial No.:	12/201,595	Group Art Unit:	1797
Filed:	AUGUST 29, 2008	Docket No.:	16291.3USD1
		Confirmation No.	9735
Title:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY		

EFS Web Transmission

PETITION FOR EXTENSION OF TIME

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

Dear Sir:

In accordance with the provisions of 37 C.F.R. §1.136(a), it is respectfully requested that a three-month extension of time be granted in which to respond to the communication mailed April 16, 2009, the period of response being extended from July 16, 2009 to October 16, 2009.

Payment is being made via credit card in the amount of $555 to cover the required extension fee for a small entity. Please charge any additional fees or credit any overpayment to Deposit Account. No. 13-2725.

Respectfully submitted,

23552
PATENT TRADEMARK OFFICE

MERCHANT & GOULD P.C.
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Minneapolis, Minnesota 55402-0903
(612) 332-5300

Dated: October 16, 2009

By
Dennis R. Daley
Reg. No. 34,994
DRD/mls

PTO/SB/06 (07-06)
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PATENT APPLICATION FEE DETERMINATION RECORD

Substitute for Form PTO-875

Application or Docket Number	Filing Date	☐ To be Mailed
12/201,595	08/29/2008	

APPLICATION AS FILED – PART I

FOR	(Column 1) NUMBER FILED	(Column 2) NUMBER EXTRA	SMALL ENTITY ☒ RATE ($)	SMALL ENTITY ☒ FEE ($)	OR	OTHER THAN SMALL ENTITY RATE ($)	OTHER THAN SMALL ENTITY FEE ($)
☐ BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A	N/A			N/A	
☐ SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A	N/A			N/A	
☐ EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A	N/A			N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	minus 20 =	*	X $ =		OR	X $ =	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	minus 3 =	*	X $ =			X $ =	
☐ APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $250 ($125 for small entity) for each additional 50 sheets or fraction thereof. See 35 U.S.C. 41(a)(1)(G) and 37 CFR 1.16(s).						
☐ MULTIPLE DEPENDENT CLAIM PRESENT (37 CFR 1.16(j))							
* If the difference in column 1 is less than zero, enter "0" in column 2.			TOTAL			TOTAL	

APPLICATION AS AMENDED – PART II

AMENDMENT 10/16/2009	(Column 1) CLAIMS REMAINING AFTER AMENDMENT		(Column 2) HIGHEST NUMBER PREVIOUSLY PAID FOR	(Column 3) PRESENT EXTRA	SMALL ENTITY RATE ($)	SMALL ENTITY ADDITIONAL FEE ($)	OR	OTHER THAN SMALL ENTITY RATE ($)	OTHER THAN SMALL ENTITY ADDITIONAL FEE ($)
Total (37 CFR 1.16(i))	* 10	Minus	** 33	= 0	X $26 =	0	OR	X $ =	
Independent (37 CFR 1.16(h))	* 1	Minus	***3	= 0	X $110 =	0	OR	X $ =	
☐ Application Size Fee (37 CFR 1.16(s))									
☐ FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))							OR		
					TOTAL ADD'L FEE	0	OR	TOTAL ADD'L FEE	

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

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

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

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

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

Legal Instrument Examiner:
/ADRIENE D. SELLMAN/

This collection of information is required by 37 CFR 1.16. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
12/201,595	08/29/2008	Edwin S. Olson	2380-00207	9735

23505 7590 04/16/2009

CONLEY ROSE, P.C.
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HOUSTON, TX 77253-3267

EXAMINER	
ORLANDO, AMBER ROSE	
ART UNIT	**PAPER NUMBER**
1797	

NOTIFICATION DATE	DELIVERY MODE
04/16/2009	ELECTRONIC

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

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

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

pathou@conleyrose.com

Office Action Summary	Application No.	Applicant(s)	
	12/201,595	OLSON ET AL.	
	Examiner	Art Unit	
	AMBER ORLANDO	1797	

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE *3* MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

1)☒ Responsive to communication(s) filed on *29 August 2008*.

2a)☐ This action is **FINAL**. 2b)☒ This action is non-final.

3)☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims

4)☒ Claim(s) *1-33* is/are pending in the application.

4a) Of the above claim(s) _____ is/are withdrawn from consideration.

5)☐ Claim(s) _____ is/are allowed.

6)☒ Claim(s) *1-33* is/are rejected.

7)☒ Claim(s) *21* is/are objected to.

8)☐ Claim(s) _____ are subject to restriction and/or election requirement.

Application Papers

9)☐ The specification is objected to by the Examiner.

10)☒ The drawing(s) filed on *29 August 2008* is/are: a)☒ accepted or b)☐ objected to by the Examiner.

Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).

Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).

11)☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

Priority under 35 U.S.C. § 119

12)☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

a)☐ All b)☐ Some * c)☐ None of:

1.☐ Certified copies of the priority documents have been received.

2.☐ Certified copies of the priority documents have been received in Application No. _____.

3.☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

* See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

1) ☒ Notice of References Cited (PTO-892)
2) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
3) ☐ Information Disclosure Statement(s) (PTO/SB/08) Paper No(s)/Mail Date _____.
4) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date. _____ .
5) ☐ Notice of Informal Patent Application
6) ☐ Other: _____.

DETAILED ACTION

Claim Objections

1. Claim 21 is objected to under 37 CFR 1.75(c), as being of improper dependent form for failing to further limit the subject matter of a previous claim. Applicant is required to cancel the claim(s), or amend the claim(s) to place the claim(s) in proper dependent form, or rewrite the claim(s) in independent form. The monitoring of the mercury content within the gas stream has nothing to do with the method of preparing a promoted carbon sorbent.

Claim Rejections - 35 USC § 112

1. The following is a quotation of the second paragraph of 35 U.S.C. 112:

> The specification shall conclude with one or more claims particularly pointing out and distinctly claiming the subject matter which the applicant regards as his invention.

Claims 4 and 24 are rejected under 35 U.S.C. 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention.

2. Claims 4 and 24 recite the limitation "the structure" in line 1 (of both). There is insufficient antecedent basis for this limitation in the claim.

Claim Rejections - 35 USC § 102

3. The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

> A person shall be entitled to a patent unless –
>
> (a) the invention was known or used by others in this country, or patented or described in a printed publication in this or a foreign country, before the invention thereof by the applicant for a patent.

4. Claims 1, 17, 18 and 20-22 are rejected under 35 U.S.C. 102(a) as being anticipated by Srinivasachar et al. US 6,848,374.

5. For claim 1, the Srinivasachar et al. reference discloses a promoted carbon sorbent comprising a base activated carbon that has been treated with a promoter selected from the group consisting of halides, halogens, and combinations thereof stream (column 6, lines 2-20). The reference does not explicitly state the carbon sorbent and the promoter reacting. It would have been obvious to one having ordinary skill in the art at the time the invention was made that contacting the activated carbon with the promoter would inherently produce the same results as that in the present invention (e.g. cause a reaction between the two). The sorbent being effective for the removal of mercury from a gas stream located downstream of a boiler, said removal of mercury yielding a cleaned gas stream, and wherein the composition of the promoted carbon sorbent is controlled by the mercury content of the cleaned gas stream is the method of using the promoted carbon. The product disclosed by the Srinivasachar et al. reference would be able to be effective for the removal of mercury from a gas stream located downstream of a boiler, said removal of mercury yielding a cleaned gas stream, and wherein the composition of the promoted carbon sorbent is controlled by the mercury content of the cleaned gas stream is the method of using the promoted carbon. Therefore the method of using the product does not further limit the claim.

6. For claim 17, the Srinivasachar reference discloses a method of preparing a promoted carbon sorbent comprising providing an activated carbon; and forming a

product by contacting 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 (column 6, lines 2-20). The reference does not explicitly state the carbon sorbent and the promoter reacting. It would have been obvious to one having ordinary skill in the art at the time the invention was made that contacting the activated carbon with the promoter would inherently produce the same results as that in the present invention (e.g. cause a reaction between the two). The sorbent being effective for the removal of mercury from a gas stream located downstream of a boiler, said removal of mercury yielding a cleaned gas stream, and wherein the composition of the promoted carbon sorbent is controlled by the mercury content of the cleaned gas stream is the method of using the promoted carbon. The product disclosed by the Srinivasachar et al. reference would be able to be effective for the removal of mercury from a gas stream located downstream of a boiler, said removal of mercury yielding a cleaned gas stream, and wherein the composition of the promoted carbon sorbent is controlled by the mercury content of the cleaned gas stream is the method of using the promoted carbon. Therefore the method of using the product does not further limit the claim. The promoted carbon sorbent being effective for removal of mercury from a gas stream downstream of a boiler, thus yielding a cleaned gas stream and wherein the composition of the sorbent is adjusted based upon the mercury content of the cleaned gas stream.

For claim 18, the claim does not point out a specific aspect of the method of preparing the promoted carbon. The claim merely states that the carbon particle has a

size (which all particles do). The claim goes on to state how the carbon promoter would be removed from a gas stream, which is the method of using the promoted carbon. The promoted carbon of the Srinivasachar reference would be able to be separated by physical means from the gas stream, and the method of using the promoted carbon does not further limit the claim.

For claim 20, the Srinivasachar et al. reference discloses preparing the sorbent in-flight by reacting the activated carbon and the promoter (column 2, lines 2-20). The reference does not explicitly state an optional secondary component. It would have been obvious to one having ordinary skill in the art at the time the invention was made to have included an additional (e.g. secondary) component comprising a halogen because the reference already established that the mercury capacity is enhanced by adding a halogen (column 6, lines 32-39) and therefore one could assume that by adding another component of the same composition would further promote the removal of mercury from the gas stream. The examiner notes that this claim recites that the secondary component is "optional" and therefore does not further limit the claim because the secondary component is not necessarily included.

For claim 21, the claim merely states the method of monitoring the mercury content within the gas stream. There is nothing with in this claim pertaining to the promoted carbon, and therefore does not further limit the claim.

For claim 22, the Srinivasachar et al. reference discloses the promoted carbon sorbent prepared by the method of claim 21 as discloses above.

7.

Claim Rejections - 35 USC § 103

The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

> (a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negatived by the manner in which the invention was made.

The factual inquiries set forth in *Graham* **v.** *John Deere Co.*, 383 U.S. 1, 148 USPQ 459 (1966), that are applied for establishing a background for determining obviousness under 35 U.S.C. 103(a) are summarized as follows:

1. Determining the scope and contents of the prior art.
2. Ascertaining the differences between the prior art and the claims at issue.
3. Resolving the level of ordinary skill in the pertinent art.
4. Considering objective evidence present in the application indicating obviousness or nonobviousness.

This application currently names joint inventors. In considering patentability of the claims under 35 U.S.C. 103(a), the examiner presumes that the subject matter of the various claims was commonly owned at the time any inventions covered therein were made absent any evidence to the contrary. Applicant is advised of the obligation under 37 CFR 1.56 to point out the inventor and invention dates of each claim that was not commonly owned at the time a later invention was made in order for the examiner to consider the applicability of 35 U.S.C. 103(c) and potential 35 U.S.C. 102(e), (f) or (g) prior art under 35 U.S.C. 103(a).

Claims 25-27, 29-31 and 33 are rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374.

For claim 25, the Srinivasachar et al. does not disclose the ratio of activated carbon to the promoter, and is chosen for optimization economic removal of mercury from the gas stream.

It would have been obvious to one having ordinary skill in the art at the time the invention was made to include the ratio of activated carbon to the promoter, and is chosen for optimization economic removal of mercury from the gas stream, since it has been held that discovering an optimum value of a result effective variable involves only routine skill in the art. *In re Boesch,* 617 F.2d 272, 205 USPQ 215 (CCPA 1980).

For claim 26, the Srinivasachar et al. reference does not explicitly state the reaction product comprises from about 1 to about 30 grams promoter per 100 grams activated carbon.

It would have been obvious to one having ordinary skill in the art at the time the invention was made to include the reaction product comprises from about 1 to about 30 grams promoter per 100 grams activated carbon, since it has been held that where the general conditions of a claim are disclosed in the prior art, discovering the optimum or workable ranges involves only routine skill in the art. *In re Aller,* 105 USPQ 233.

For claim 27, the Srinivasachar et al. reference does not disclose the promoter is in an organic solvent when contacting the activated carbon. The reference does disclose putting the promoter into a solvent in order to be able to vaporize the promoter (column 6, lines 2-20). It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the reference to include

finding any type of solvent (including organic) to dissolve the promoter because this allows the promoter to be vaporized and have better contact with the activated carbon.

For claim 29, the Srinivasachar et al. reference discloses the promoter is in the gas phase when contacting the activated carbon (column 6, lines 2-20).

For claim 30, the Srinivasachar et al. reference discloses the promoter is selected from the group consisting of Br_2, HBr, and combinations thereof (column 6, lines 9-10).

For claim 31, the Srinivasachar et al. discloses reacting the 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 (column 6, lines 2-20). The reference does not explicitly state the component being the secondary component. It would have been obvious to one having ordinary skill in the art at the time the invention was made to have included and additional (e.g. secondary) component comprising a halogen because the reference already established that the mercury capacity is enhanced by adding a halogen (column 6, lines 2-20) and therefore one could assume that by adding another component of the same composition would further promote the removal of mercury from the gas stream. The examiner notes that this claim recites that the secondary component is "optional" and therefore does not further limit the claim because the secondary component is not necessarily included.

For claim 33 the Srinivasachar reference discloses combining the promoter and the activated carbon (column 6, lines 2-20). The reference does not disclose injecting the optional secondary component and the promoter simultaneously. The promoters are

essentially the same so therefore it would be obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar reference to include disclose injecting the optional secondary component and the promoter simultaneously because this will cut down on production costs.

Claim 2 is rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374 as applied in claim 1 above, and further in view of Reiss 5,575,982.

For claim 2, the Srinivasachar et al. reference discloses 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 (column 1, lines 37-45 and column 5, lines 20-29) and the promoter is selected from the group consisting of molecular halogens, Group V halides, Group VI halides, hydrohalides, and combinations thereof (column 6, lines 2-20). The reference does not disclose an average particle size greater than 40 micrometers

The Reiss reference discloses the activated carbon has an average particle size greater than 40 μm (Reiss, column 4, lines 59-65).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar reference to include the activated carbon has an average particle size greater than 40 μm (Reiss column 4, lines 59-65) because these particle diameters of the activated carbon can be effectively introduced into the exhaust gas stream and cause optimum adsorption.

Claims 3-6 are rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374 and Reiss 5,575,982 as applied in claim 2 above, and further in view of Moller et al. US 4,889,698.

For claims 3 and 4, the Srinivasachar et al. reference does not disclose the sorbent comprises an activated carbon matrix in a fixed or a moving bed structure and the structure is selected from the group consisting of moving fiber screens and monoliths.

The Moller et al. reference discloses using the sorbent comprises an activated carbon matrix in a fixed or a moving bed structure (column 4, lines 39-45). The reference does not explicitly state the structure is selected from the group consisting of moving fiber screens and monoliths. Merely choosing different types of fixed bed structures would be obvious to one having ordinary skill in the art.

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar et al. reference to include the sorbent comprises an activated carbon matrix in a fixed or a moving bed structure and the structure is selected from the group consisting of moving fiber screens and monoliths (Moller et al. column 4, lines 39-45) because this allows the contaminated gas to reach as much of the activated carbon as possible.

For claim 5, the Srinivasachar et al. reference does not disclose an optional alkaline component selected from the group consisting of alkali elements, alkaline earth elements, alkali salts, alkaline earth salts, and combinations thereof. The examiner notes that this claim recites that the alkaline component is "optional" and therefore does

not further limit the claim because the alkaline component is not necessarily included. Furthermore the claim does not state how the alkaline substance is included within the promoted carbon sorbent (e.g. is it reacting/bonding/coating). If it is not reacting/bonding/coating, then it is not part of the promoted carbon sorbent and does not further limit the claim.

The Moller et al. reference discloses an optional alkaline component selected from the group consisting of alkali elements, alkaline earth elements, alkali salts, alkaline earth salts, and combinations thereof (column 7, lines 43-56).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar et al. reference to include an optional alkaline component selected from the group consisting of alkali elements, alkaline earth elements, alkali salts, alkaline earth salts, and combinations thereof (Moller et al. column 7, lines 43-56) because this promotes the removal of mercury from the gas.

For claim 6, neither the Srinivasachar et al. nor the Moller et al. references discloses the ratio of base activated carbon, the promoter, and the optional alkaline component is chosen for optimum economic removal of mercury from the gas stream.

It would have been obvious to one having ordinary skill in the art at the time the invention was made to include the ratio of base activated carbon, the promoter, and the optional alkaline component is chosen for optimum economic removal of mercury from the gas stream, since it has been held that discovering an optimum value of a result

effective variable involves only routine skill in the art. *In re Boesch,* 617 F.2d 272, 205 USPQ 215 (CCPA 1980).

Claims 7 and 8 are rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374 and Reiss 5,575,982 as applied in claim 2 above, and further in view of Broderick US 6,942,840.

For claim 7, the Srinivasachar et al. reference does not disclose 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.

The Broderick reference discloses disclose 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 (column 4, lines 12-25).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar et al. reference to include disclose 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 (Broderick column 4, lines 12-25) because this allows the mercury to vented to the atmosphere without additional treatment.

For claim 8 the Srinivasachar et al. reference discloses the promoter is selected from the group consisting of Br_2, HBr, and combinations thereof (column 6, lines 9-10).

Claim 9 is rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374, Reiss 5,575,982 and Broderick US 6,942,840 as applied in claim 7 above, and further in view of Moller et al. US 4,889,698.

For claims 9, the Srinivasachar et al. reference discloses the promoter is selected from the group consisting of Br_2, HBr, and combinations thereof (column 6, lines 9-10). The reference does not disclose an optional alkaline component comprising an alkaline or an alkaline earth element. The examiner notes that this claim recites that the alkaline component is "optional" and therefore does not further limit the claim because the alkaline component is not necessarily included. Furthermore the claim does not state how the alkaline substance is included within the promoted carbon sorbent (e.g. is it reacting/bonding/coating). If it is not reacting/bonding/coating, then it is not part of the promoted carbon sorbent and does not further limit the claim.

The Moller et al. reference discloses an optional alkaline component selected from the group consisting of alkali elements, alkaline earth elements, alkali salts, alkaline earth salts, and combinations thereof (column 7, lines 43-56).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar et al. reference to include an optional alkaline component selected from the group consisting of alkali elements, alkaline earth elements, alkali salts, alkaline earth salts, and combinations thereof (Moller et al. column 7, lines 43-56) because this promotes the removal of mercury from the gas.

Claims 10-16 are rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374 and Reiss 5,575,982 as applied in claim 2 above, and further in view of Moller et al. US 4,889,698.

For claims 10 and 12-14, the Srinivasachar et al. discloses an optional component comprising a halogen or a hydrohalide such that the reactivity and mercury capacity of the sorbent are enhanced, the optional component is selected from the group consisting of Group V halides, Group VI halides, HI, HBr, HCl, and combinations thereof, and the promoter is substantially in vapor form when combined with teh base activated carbon (column 6, lines 2-20). The reference does not explicitly state the component being the secondary component. It would have been obvious to one having ordinary skill in the art at the time the invention was made to have included an additional (e.g. secondary) component comprising a halogen because the reference already established that the mercury capacity is enhanced by adding a halogen (column 6, lines 32-39) and therefore one could assume that by adding another component of the same composition would further promote the removal of mercury from the gas stream. The examiner notes that this claim recites that the secondary component is "optional" and therefore does not further limit the claim because the secondary component is not necessarily included. Furthermore the claim does not state how the secondary component is included within the promoted carbon sorbent (e.g. is it reacting/bonding/coating). If it is not reacting/bonding/coating, then it is not part of the promoted carbon sorbent and does not further limit the claim.

The Moller et al. reference discloses an optional alkaline component selected from the group consisting of alkali elements, alkaline earth elements, alkali salts, alkaline earth salts, and combinations thereof (column 7, lines 43-56).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar et al. reference to include an optional alkaline component selected from the group consisting of alkali elements, alkaline earth elements, alkali salts, alkaline earth salts, and combinations thereof (Moller et al. column 7, lines 43-56) because this promotes the removal of mercury from the gas.

For claim 11, the Srinivasachar et al. reference does not explicitly state the concentration of the optional secondary component on the finished sorbent is within the range of from about 1 wt% to about 15 wt% of the concentration of the promoter on the finished sorbent.

It would have been obvious to one having ordinary skill in the art at the time the invention was made to include the concentration of the optional secondary component on the finished sorbent is within the range of from about 1 wt% to about 15 wt% of the concentration of the promoter on the finished sorbent, since it has been held that where the general conditions of a claim are disclosed in the prior art, discovering the optimum or workable ranges involves only routine skill in the art. *In re Aller,* 105 USPQ 233.

For claim 15, the Srinivasachar et al. reference does not disclose the promoter is combined with an organic solvent prior to reaction with the base activated carbon. The reference does disclose putting the promoter into a solvent in order to be able to vaporize the promoter (column 6, lines 2-20). It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the reference to include finding any type of solvent (including organic) to dissolve the

promoter because this allows the promoter to be vaporized and have better contact with the activated carbon.

For claim 16 the Srinivasachar reference discloses combining the promoter and the activated carbon (column 6, lines 2-20). The reference does not disclose injecting the optional secondary component and the promoter simultaneously. The promoters are essentially the same so therefore it would be obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar reference to include injecting the optional secondary component and the promoter simultaneously because this will cut down on production costs.

Claims 19 is rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374 as applied in claim 18 above, and further in view of Reiss 5,575,982.

For claim 19, the Srinivasachar reference does not disclose the reaction product has an average particle size distribution greater than about 40 micrometers.

The Reiss reference discloses the activated carbon has an average particle size greater than 40 μm (Reiss, column 4, lines 59-65).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar reference to include the activated carbon has an average particle size greater than 40 μm (Reiss column 4, lines 59-65) because these particle diameters of the activated carbon can be effectively introduced into the exhaust gas stream and cause optimum adsorption.

Claims 23 and 24 are rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374 as applied in claim 17 above, and further in view of Moller et al. US 4,889,698.

For claims 23 and 24, the Srinivasachar et al. reference does not disclose the sorbent comprises an activated carbon matrix in a fixed or a moving bed structure and the structure is selected from the group consisting of moving fiber screens and monoliths.

The Moller et al. reference discloses using the sorbent comprises an activated carbon matrix in a fixed or a moving bed structure (column 4, lines 39-45). The reference does not explicitly state the structure is selected from the group consisting of moving fiber screens and monoliths. Merely choosing different types of fixed bed structures would be obvious to one having ordinary skill in the art.

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar et al. reference to include the sorbent comprises an activated carbon matrix in a fixed or a moving bed structure and the structure is selected from the group consisting of moving fiber screens and monoliths (Moller et al. column 4, lines 39-45) because this allows the contaminated gas to reach as much of the activated carbon as possible.

Claim 28 is rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374 as applied in claim 17 above, and further in view of Moller et al. US 4,889,698.

For claim 28 the Srinivasachar et al. reference does not disclose providing an optional alkaline component to improve the effectiveness of mercury removal. The examiner notes that this claim recites that the alkaline component is "optional" and therefore does not further limit the claim because the alkaline component is not necessarily included. Furthermore the claim does not state how the alkaline substance is included within the promoted carbon sorbent (e.g. is it reacting/bonding/coating). If it is not reacting/bonding/coating, then it is not part of the promoted carbon sorbent and does not further limit the claim.

The Moller et al. reference discloses providing an optional alkaline component to improve the effectiveness of mercury removal (column 7, lines 43-56).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar et al. reference to include providing an optional alkaline component to improve the effectiveness of mercury removal (Moller et al. column 7, lines 43-56) because this promotes the removal of mercury from the gas.

Claim 32 is rejected under 35 U.S.C. 103(a) as being unpatentable over Srinivasachar et al. US 6,848,374 as applied in claim 31 above, and further in view of Moller et al. US 4,889,698.

For claim 32, the Srinivasachar et al. reference does not disclose reacting the 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.

The Moller et al. reference discloses reacting the 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 (column 7, lines 43-56).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Srinivasachar et al. reference to include reacting the 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 (Moller et al. column 7, lines 43-56) because this promotes the removal of mercury from the gas.

Conclusion

Any inquiry concerning this communication or earlier communications from the examiner should be directed to AMBER ORLANDO whose telephone number is (571)270-3149. The examiner can normally be reached on Mon.-Thurs. (6:30-5:00).

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Walter Griffin can be reached on (571) 272-1447. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

AO

/Walter D. Griffin/
Supervisory Patent Examiner, Art Unit 1797

Notice of References Cited	Application/Control No. 12/201,595	Applicant(s)/Patent Under Reexamination OLSON ET AL.	
	Examiner AMBER ORLANDO	Art Unit 1797	Page 1 of 1

U.S. PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Name	Classification
*	A	US-4,820,318 A	04-1989	Chang et al.	95/141
*	B	US-4,889,698 A	12-1989	Moller et al.	423/210
*	C	US-5,300,137 A	04-1994	Weyand et al.	75/670
*	D	US-5,409,522 A	04-1995	Durham et al.	75/670
*	E	US-5,575,982 A	11-1996	Reiss et al.	423/210
*	F	US-6,372,187 B1	04-2002	Madden et al.	422/171
*	G	US-6,375,909 B1	04-2002	Dangtran et al.	423/235
*	H	US-2004/0013589 A1	01-2004	Vosteen et al.	423/210
*	I	US-6,818,043 B1	11-2004	Chang et al.	95/37
*	J	US-6,848,374 B2	02-2005	Srinivasachar et al.	110/345
*	K	US-6,942,840 B1	09-2005	Broderick, Thomas E.	423/101
*	L	US-2006/0048646 A1	03-2006	Olson et al.	095/134
	M	US-			

FOREIGN PATENT DOCUMENTS

*		Document Number Country Code-Number-Kind Code	Date MM-YYYY	Country	Name	Classification
	N					
	O					
	P					
	Q					
	R					
	S					
	T					

NON-PATENT DOCUMENTS

*		Include as applicable: Author, Title Date, Publisher, Edition or Volume, Pertinent Pages)
	U	
	V	
	W	
	X	

*A copy of this reference is not being furnished with this Office action. (See MPEP § 707.05(a).)
Dates in MM-YYYY format are publication dates. Classifications may be US or foreign.

EAST Search History

Ref #	Hits	Search Query	DBs	Default Operator	Plurals	Time Stamp
L1	135	carbon with alkali same remov$4 with mercury	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 16:18
L2	55234	carbon with alkali (salts or elements) same remov$4 with mercury	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 16:18
L3	2	carbon with alkali (salts or elements) same remov$4 with mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:19
L4	135	carbon with alkali same remov$4 with mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:19
L5	15	carbon with alkali same remov$4 with mercury and exhaust	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:19
L6	4	mercury with exhaust and mercury with stabili$5 with reagent and carbon with remov $4 with mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:29
L7	6	"6848374" and (stabili$4 or sulfur or sulfide)	US-PGPUB; USPAT; USOCR	ADJ	ON	2009/04/01 16:31
L8	0	mercury with exhaust and mercury with stabili$5 with (hydrogen sulfide or sulfer dioxide) and carbon with remov$4 with mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:34

L9	0	mercury with exhaust and mercury with stabili$5 with (hydrogen sulfide or sulfur dioxide) and carbon with remov$4 with mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:34
L10	2	stabili$5 with (hydrogen sulfide or sulfur dioxide) and carbon with remov$4 with mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:35
L11	2	mercury with exhaust and mercury with stabili$5 with sulfur and carbon with remov$4 with mercury	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 16:36
L12	4	"6848374" and (organic or solvent)	US-PGPUB; USPAT; USOCR	ADJ	ON	2009/04/01 16:46
L13	5	"6848374" and (organic or solvent or dissolv $4)	US-PGPUB; USPAT; USOCR	ADJ	ON	2009/04/01 16:46
L14	7	carbon with alkali same remov$4 with mercury and (fixed or moving) bed	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 17:21
L15	8	carbon with halogen same remov$4 with mercury and (fixed or moving) bed	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 17:24
L16	5	carbon with halogen same remov$4 with mercury and (monolith or fiber screens)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 17:30

L17	20	activated carbon same remov$4 with mercury and (monolith or fiber screens)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 17:31
L18	0	activated carbon same remov$4 with mercury and activated carbong with (monolith or fiber screens)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 17:32
L19	2	activated carbon same remov$4 with mercury and activated carbon with (monolith or fiber screens)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 17:32
L20	2	activated carbon same remov$4 with mercury and activated carbon with (monolith or fiber screen)	US-PGPUB; USPAT; USOCR	ADJ	OFF	2009/04/01 17:32
L21	67	"4,889,698"	US-PGPUB; USPAT; USOCR	ADJ	ON	2009/04/01 17:36
L22	125	95/134.ccls. and remov$4 with mercury	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 19:25
L23	20	95/134.ccls. and remov$4 with mercury and halogen	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 19:26
L24	3	95/142.ccls. and remov$4 with mercury and carbon and halide	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 19:26
L25	0	11//345.ccls. and remov$4 with mercury and carbon and (halide or halogen)	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 19:28

L26	8	110/345.ccls. and remov$4 with mercury and carbon and (halide or halogen)	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 19:28
L27	54	423/210.ccls. and remov$4 with mercury and carbon and (halide or halogen)	US-PGPUB; USPAT; USOCR	OR	OFF	2009/04/01 19:28

4/1/2009 7:34:01 PM
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Search Notes	Application/Control No.	Applicant(s)/Patent Under Reexamination
	12201595	OLSON ET AL.
	Examiner	Art Unit
	AMBER ORLANDO	1797

SEARCHED

Class	Subclass	Date	Examiner
110	345	4/1/2009	AO
423	210	4/1/2009	AO
95	134, 142	4/1/2009	AO

SEARCH NOTES

Search Notes	Date	Examiner
Inventor Search (eDAN)	4/1/2009	AO
Class 95 searched for a carbon sorbent having a promoter. (EAST)	4/1/2009	AO
Classes 423 and 110 searched for a carbon sorbent having a halide promoter and an alkaline component. (EAST)	4/1/2009	AO

INTERFERENCE SEARCH

Class	Subclass	Date	Examiner

/AMBER ORLANDO/ Acting Examiner.Art Unit 1797	

Index of Claims	Application/Control No.	Applicant(s)/Patent Under Reexamination
	12201595	OLSON ET AL.
	Examiner	Art Unit
	AMBER ORLANDO	1797

✓	Rejected	-	Cancelled	N	Non-Elected	A	Appeal
=	Allowed	÷	Restricted	I	Interference	O	Objected

☐ Claims renumbered in the same order as presented by applicant ☐ CPA ☐ T.D. ☐ R.1.47

CLAIM		DATE								
Final	Original	04/01/2009	04/01/2009							
	1	✓								
	2	✓								
	3	✓								
	4	✓								
	5	✓								
	6	✓								
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BIB DATA SHEET

CONFIRMATION NO. 9735

SERIAL NUMBER	FILING or 371(c) DATE RULE	CLASS	GROUP ART UNIT	ATTORNEY DOCKET NO.
12/201,595	08/29/2008	095	1797	2380-00207

APPLICANTS
Edwin S. Olson, Grand Forks, ND;
Michael J. Holmes, Thompson, ND;
John H. Pavlish, East Grand Forks, MN;

**** CONTINUING DATA ***************************
This application is a DIV of 11/209,163 08/22/2005 PAT 7,435,286
which claims benefit of 60/605,640 08/30/2004

**** FOREIGN APPLICATIONS ***************************

**** IF REQUIRED, FOREIGN FILING LICENSE GRANTED **** ** SMALL ENTITY **
09/10/2008

Foreign Priority claimed ☐ Yes ☑ No 35 USC 119(a-d) conditions met ☐ Yes ☑ No Verified and Acknowledged /AMBER ROSE ORLANDO/ Examiner's Signature	☐ Met after Allowance Initials	STATE OR COUNTRY	SHEETS DRAWINGS	TOTAL CLAIMS	INDEPENDENT CLAIMS
		ND	7	33	2

ADDRESS

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David A. Rose
P. O. BOX 3267
HOUSTON, TX 77253-3267
UNITED STATES

TITLE

SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Not for submission under 37 CFR 1.99)

Application Number	12201595
Filing Date	2008-08-29
First Named Inventor	Edwin S. Olson
Art Unit	1797
Examiner Name	TBD
Attorney Docket Number	2380-00207

U.S.PATENTS

Examiner Initial*	Cite No	Patent Number	Kind Code[1]	Issue Date	Name of Patentee or Applicant of cited Document	Pages,Columns,Lines where Relevant Passages or Relevant Figures Appear
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FOREIGN PATENT DOCUMENTS

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	1	1099490	CA		1981-04-21	Geert, et al.		☐
	2	2400898	CA		2001-08-30	Maes et al.		☐

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INFORMATION DISCLOSURE STATEMENT BY APPLICANT (**Not for submission under 37 CFR 1.99)**		
	Application Number	12201595
	Filing Date	2008-08-29
	First Named Inventor	Edwin S. Olson
	Art Unit	1797
	Examiner Name	TBD
	Attorney Docket Number	2380-00207

Examiner Initials*	Cite No	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc), date, pages(s), volume-issue number(s), publisher, city and/or country where published.	T[5]
	1	Official Action from the Canadian Patent Office, dated March 3, 2009, which relates to Canadian Patent Application No. 2,584,327, filed August 23, 2005, 4 pages.	☐

If you wish to add additional non-patent literature document citation information please click the Add button

EXAMINER SIGNATURE

Examiner Signature		Date Considered	

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through a citation if not in conformance and not considered. Include copy of this form with next communication to applicant.

[1] See Kind Codes of USPTO Patent Documents at www.USPTO.GOV or MPEP 901.04. [2] Enter office that issued the document, by the two-letter code (WIPO Standard ST.3). [3] For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document. [4] Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible. [5] Applicant is to place a check mark here if English language translation is attached.

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	Application Number	12201595
	Filing Date	2008-08-29
	First Named Inventor	Edwin S. Olson
	Art Unit	1797
	Examiner Name	TBD
	Attorney Docket Number	2380-00207

CERTIFICATION STATEMENT

Please see 37 CFR 1.97 and 1.98 to make the appropriate selection(s):

☒ That each item of information contained in the information disclosure statement was first cited in any communication from a foreign patent office in a counterpart foreign application not more than three months prior to the filing of the information disclosure statement. See 37 CFR 1.97(e)(1).

OR

☐ That no item of information contained in the information disclosure statement was cited in a communication from a foreign patent office in a counterpart foreign application, and, to the knowledge of the person signing the certification after making reasonable inquiry, no item of information contained in the information disclosure statement was known to any individual designated in 37 CFR 1.56(c) more than three months prior to the filing of the information disclosure statement. See 37 CFR 1.97(e)(2).

☐ See attached certification statement.

☐ Fee set forth in 37 CFR 1.17 (p) has been submitted herewith.

☐ None

SIGNATURE

A signature of the applicant or representative is required in accordance with CFR 1.33, 10.18. Please see CFR 1.4(d) for the form of the signature.

Signature	/Timothy S. Westby/	Date (YYYY-MM-DD)	2009-04-01
Name/Print	Timothy S. Westby	Registration Number	52352

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EFS ID:	5077342
Application Number:	12201595
International Application Number:	
Confirmation Number:	9735
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Customer Number:	23505
Filer:	Timothy S. Westby
Filer Authorized By:	
Attorney Docket Number:	2380-00207
Receipt Date:	01-APR-2009
Filing Date:	29-AUG-2008
Time Stamp:	15:31:19
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	no

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1	NPL Documents	2380-00207CanadianOfficeAction.pdf	137151 5fce98a6c297a7d8c5aa8bc487dbf2657e44f336	no	4

Warnings:

Information:

2	Foreign Reference	Geert.pdf	406575 e00353ec0918e969dabf5a607f2ab3853bd08051	no	11
Warnings:					
Information:					
3	Foreign Reference	Maes.pdf	635515 72af063e49b4c6c826b47c1a52258f93ca3148bd	no	13
Warnings:					
Information:					
4	Information Disclosure Statement (IDS) Filed (SB/08)	2380-00207InformationDisclosureStatement.pdf	81699 74ae1a0ecd5620b66f5515ada9c06bb5054ec033	no	4
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Information:					
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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

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.

(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

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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(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

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

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.

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

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

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.

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.

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.

1/1

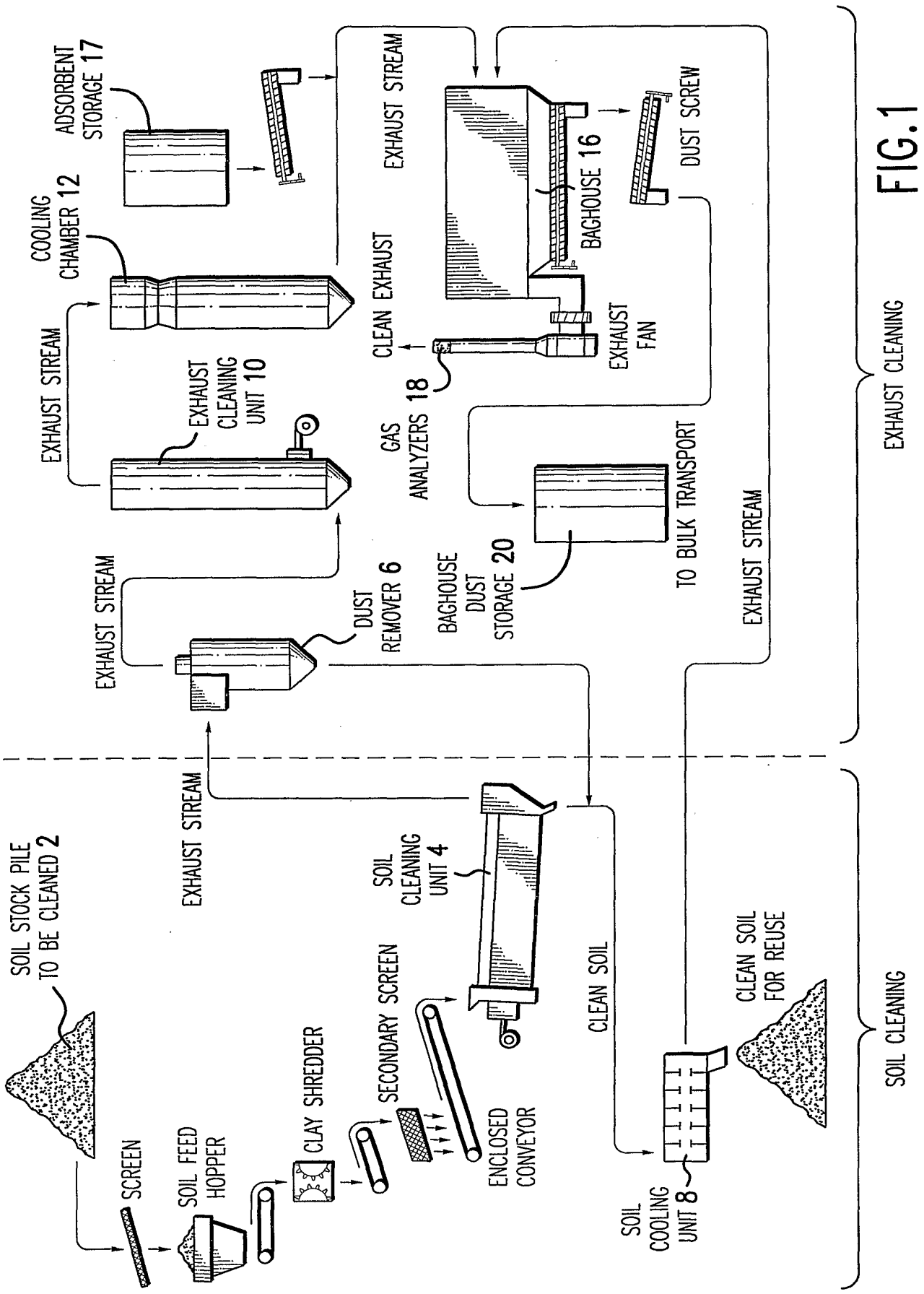

SOIL STOCK PILE TO BE CLEANED 2
SCREEN
SOIL FEED HOPPER
CLAY SHREDDER
SECONDARY SCREEN
ENCLOSED CONVEYOR
SOIL CLEANING UNIT 4
EXHAUST STREAM
DUST REMOVER 6
EXHAUST STREAM
EXHAUST CLEANING UNIT 10
EXHAUST STREAM
COOLING CHAMBER 12
ADSORBENT STORAGE 17
EXHAUST STREAM
CLEAN EXHAUST
GAS ANALYZERS 18
BAGHOUSE 16
DUST SCREW
EXHAUST FAN
BAGHOUSE DUST STORAGE 20
TO BULK TRANSPORT
EXHAUST STREAM
CLEAN SOIL
SOIL COOLING UNIT 8
CLEAN SOIL FOR REUSE
EXHAUST CLEANING
SOIL CLEANING

FIG. 1

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NOTICE OF PUBLICATION OF APPLICATION

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

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The publication process established by the Office does not provide for mailing a copy of the publication to applicant. A copy of the publication may be obtained from the Office upon payment of the appropriate fee set forth in 37 CFR 1.19(a)(1). Orders for copies of patent application publications are handled by the USPTO's Office of Public Records. The Office of Public Records can be reached by telephone at (703) 308-9726 or (800) 972-6382, by facsimile at (703) 305-8759, by mail addressed to the United States Patent and Trademark Office, Office of Public Records, Alexandria, VA 22313-1450 or via the Internet.

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

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

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APPLICATION NUMBER	FILING or 371(c) DATE	GRP ART UNIT	FIL FEE REC'D	ATTY.DOCKET.NO	TOT CLAIMS	IND CLAIMS
12/201,595	08/29/2008	1797	825	2380-00207	33	2

CONFIRMATION NO. 9735

UPDATED FILING RECEIPT

23505
CONLEY ROSE, P.C.
David A. Rose
P. O. BOX 3267
HOUSTON, TX 77253-3267

OC00000033222557

Date Mailed: 11/26/2008

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

Applicant(s)

Edwin S. Olson, Grand Forks, ND;
Michael J. Holmes, Thompson, ND;
John H. Pavlish, East Grand Forks, MN;

Assignment For Published Patent Application

ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION, Grand Forks, ND

Power of Attorney: None

Domestic Priority data as claimed by applicant

This application is a DIV of 11/209,163 08/22/2005 PAT 7,435,286
which claims benefit of 60/605,640 08/30/2004

Foreign Applications

If Required, Foreign Filing License Granted: 09/10/2008

The country code and number of your priority application, to be used for filing abroad under the Paris Convention, is **US 12/201,595**

Projected Publication Date: 03/05/2009

Non-Publication Request: No

Early Publication Request: No
** **SMALL ENTITY** **

Title

SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

Preliminary Class

095

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

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Title 37, Code of Federal Regulations, 5.11 & 5.15

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APPLICATION NUMBER	FILING OR 371(C) DATE	FIRST NAMED APPLICANT	ATTY. DOCKET NO./TITLE
12/201,595	08/29/2008	Edwin S. Olson	2380-00207

CONFIRMATION NO. 9735

FORMALITIES LETTER

23505
CONLEY ROSE, P.C.
David A. Rose
P. O. BOX 3267
HOUSTON, TX 77253-3267

OC000000032016577

CR Ref. # 2380-00207
Action Rec'd: POZIA
Due Date: 11/17/08
Other: Oath + Drawings
Docketed: ☑ yes ☐ no initials
Check'd by Sec:
Completed:

Date Mailed: 09/17/2008

NOTICE TO FILE MISSING PARTS OF NONPROVISIONAL APPLICATION

FILED UNDER 37 CFR 1.53(b)

Filing Date Granted

Items Required To Avoid Abandonment:

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

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

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

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

- Replacement drawings in compliance with 37 CFR 1.84 and 37 CFR 1.121(d) are required. The drawings submitted are not acceptable because:
  - The drawings have a line quality that is too light to be reproduced (weight of all lines and letters must be heavy enough to permit adequate reproduction) or text that is illegible (reference characters, sheet numbers, and view numbers must be plain and legible) see 37 CFR 1.84(l) and (p)(1)); See Figure(s) 2, 4-10.
  - Numbers, letters, and reference characters on the drawings must measure at least 0.32 cm (1/8 inch) in height. See Figure(s) 4-10.

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

The applicant needs to satisfy supplemental fees problems indicated below.

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

- To avoid abandonment, a surcharge (for late submission of filing fee, search fee, examination fee or oath or declaration) as set forth in 37 CFR 1.16(f) of **$65** for a small entity in compliance with 37 CFR 1.27, must be submitted with the missing items identified in this notice.

SUMMARY OF FEES DUE:

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- $65 Surcharge.

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PTO/SB/01A (08-03)
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DECLARATION (37 CFR 1.63) FOR UTILITY OR DESIGN APPLICATION USING AN APPLICATION DATA SHEET (37 CFR 1.76)

Title of Invention	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

As below named inventors, we declare that:

This Declaration is directed to:

☐ The attached application, or

☒ Application No. 12/201,595, filed on August 29, 2008.

☐ as amended on (if applicable);

We believe that we are the original and first inventors of the subject matter which is claimed and for which a patent is sought;

We have reviewed and understand the contents of the above-identified application, including the claims, as amended by any amendment specifically referred to above;

We acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to us to be material to patentability as defined in 37 CFR 1.56, including for continuation-in-part applications, material information which became available between the filing date of the prior application and the national or PCT International filing date of the continuation-in-part application.

All statements made herein of our own knowledge are true, all statements made herein on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like are punishable by fine or imprisonment, or both, under 18 U.S.C. 1001, and may jeopardize the validity of the application or any patent issuing thereon.

FULL NAME OF INVENTOR(S)

Inventor one: Edwin S. Olson — Date: 10-14, 2008
Signature: Edwin S. Olson — Citizen of: US

Inventor two: Michael J. Holmes — Date: 9/25, 2008
Signature: Michael J. Holmes — Citizen of: US

Inventor three: John H. Pavlish — Date: 9/26, 2008
Signature: John H. Pavlish — Citizen of: US

Inventor four: — Date: _____, 2008
Signature: — Citizen of:

Inventor five: — Date: _____, 2008
Signature: — Citizen of:

☐ Additional inventors are being named on additional form(s) attached hereto.

This collection of information is required by 35 U.S.C. 115 and CFR 1.63. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 1 minute to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P. O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Commissioner for Patents, P. O. Box 1450, Alexandria, VA 22313-1450.
If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

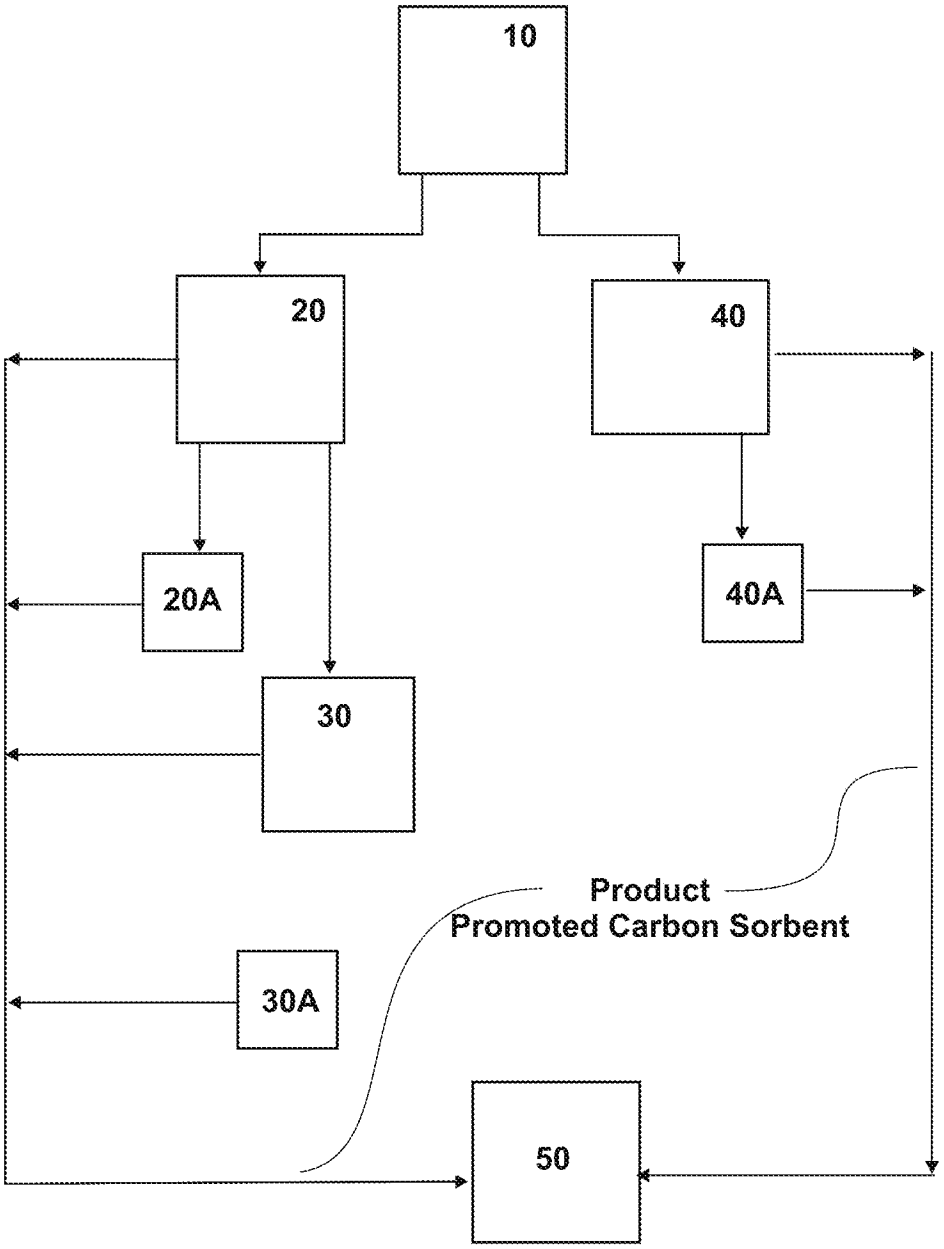


FIG. 1

2/7

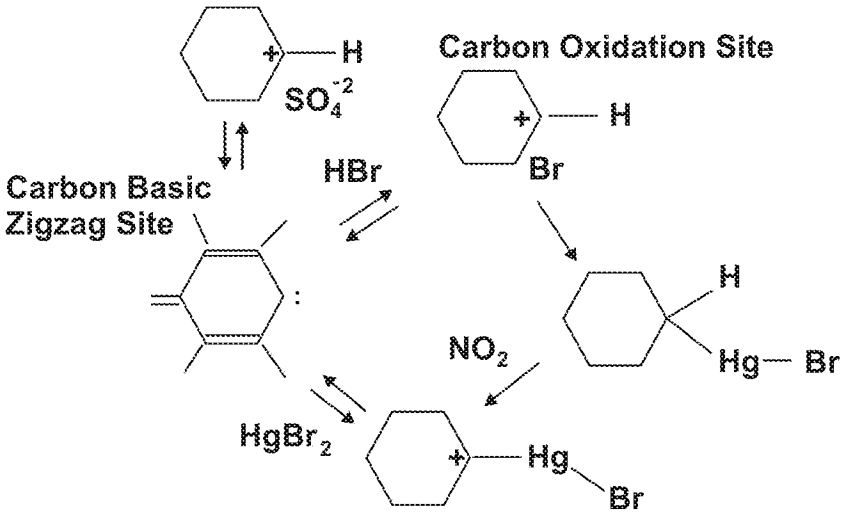

Carbon Oxidation Site
H
SO_4^{-2}
H
Br
HBr
Carbon Basic
Zigzag Site
H
Hg— Br
NO_2
$HgBr_2$
Hg
Br

FIG. 2

3/7

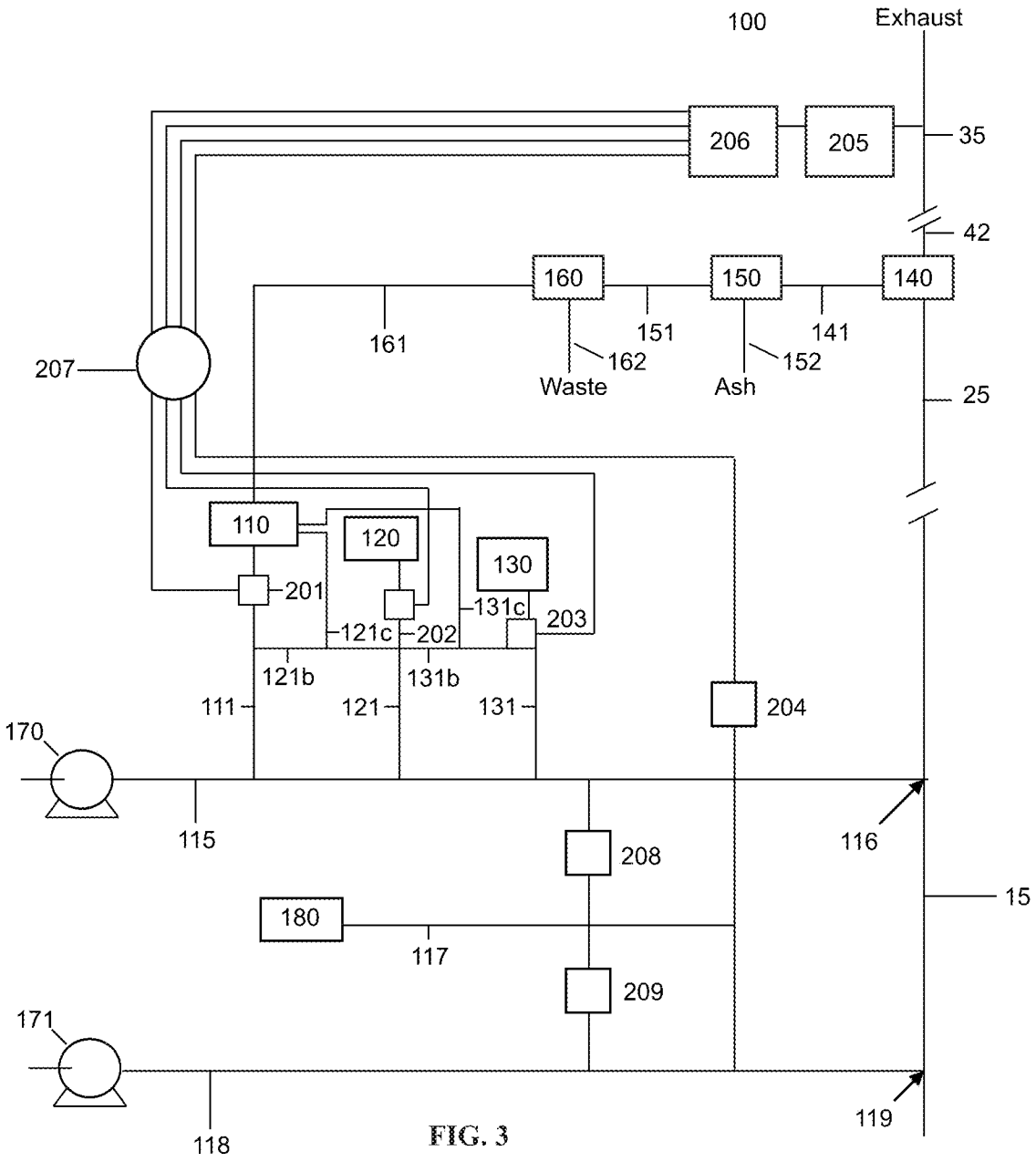

100
Exhaust
206
205
35
42
160
150
140
161
151
141
162
152
207
Waste
Ash
25
110
120
130
201
131c
203
121c
202
121b
131b
111
121
131
204
170
115
116
208
15
180
117
209
171
118
119

FIG. 3

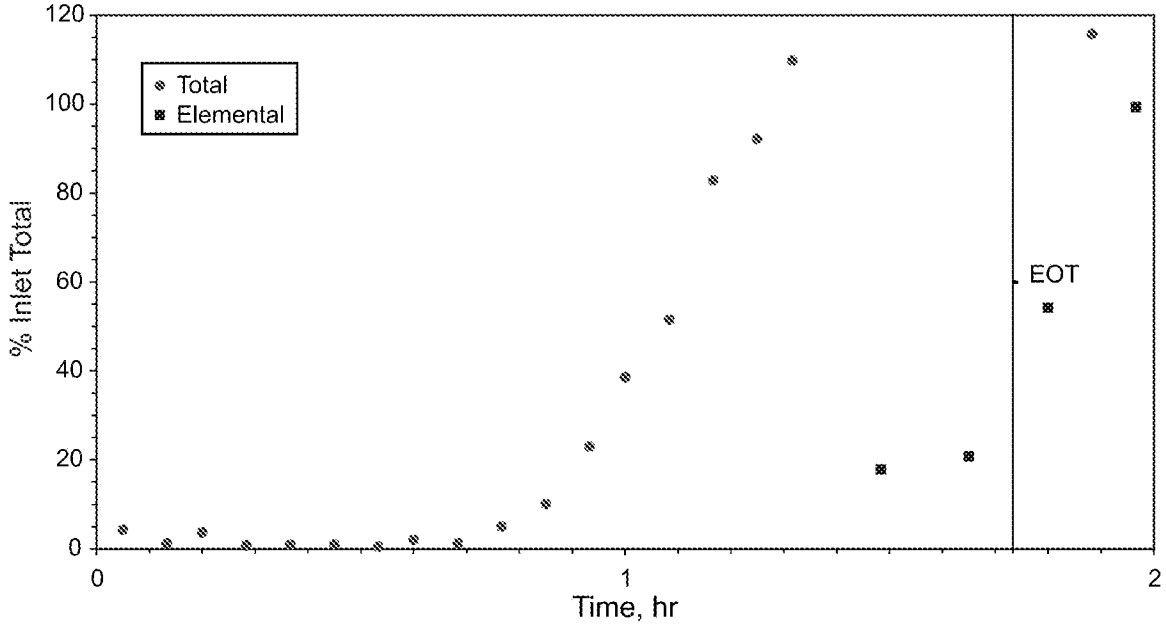

120
100
80
60
40
20
0
% Inlet Total
Total
Elemental
EOT
0
1
2
Time, hr

FIG. 4

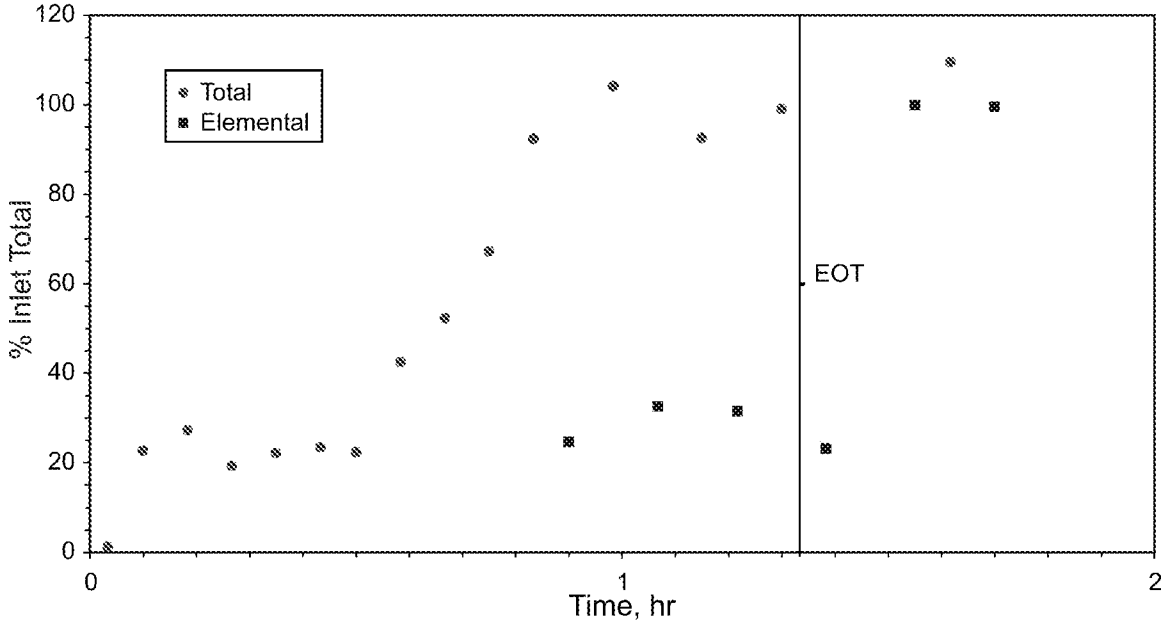

120
100
80
60
40
20
0
% Inlet Total
Total
Elemental
EOT
0
1
2
Time, hr

FIG. 5

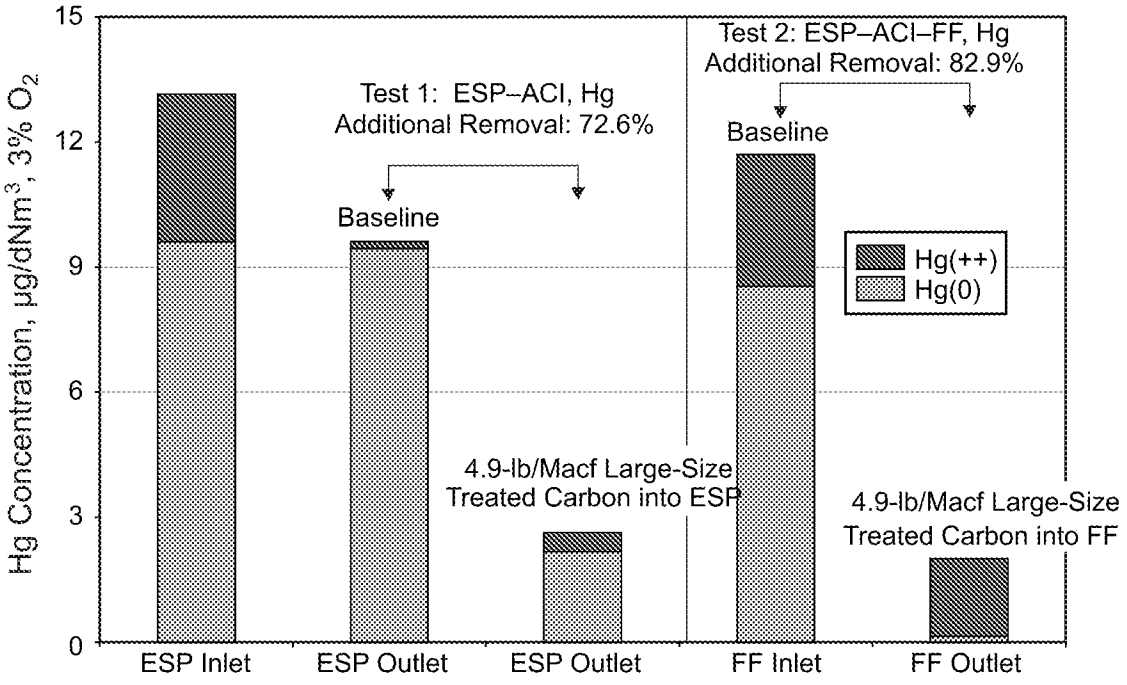

Hg Concentration, µg/dNm3, 3% O2
15
12
9
6
3
0
Test 1: ESP–ACI, Hg
Additional Removal: 72.6%
Baseline
Test 2: ESP–ACI–FF, Hg
Additional Removal: 82.9%
Baseline
Hg(++)
Hg(0)
4.9-lb/Macf Large-Size
Treated Carbon into ESP
4.9-lb/Macf Large-Size
Treated Carbon into FF
ESP Inlet
ESP Outlet
ESP Outlet
FF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

FIG. 6

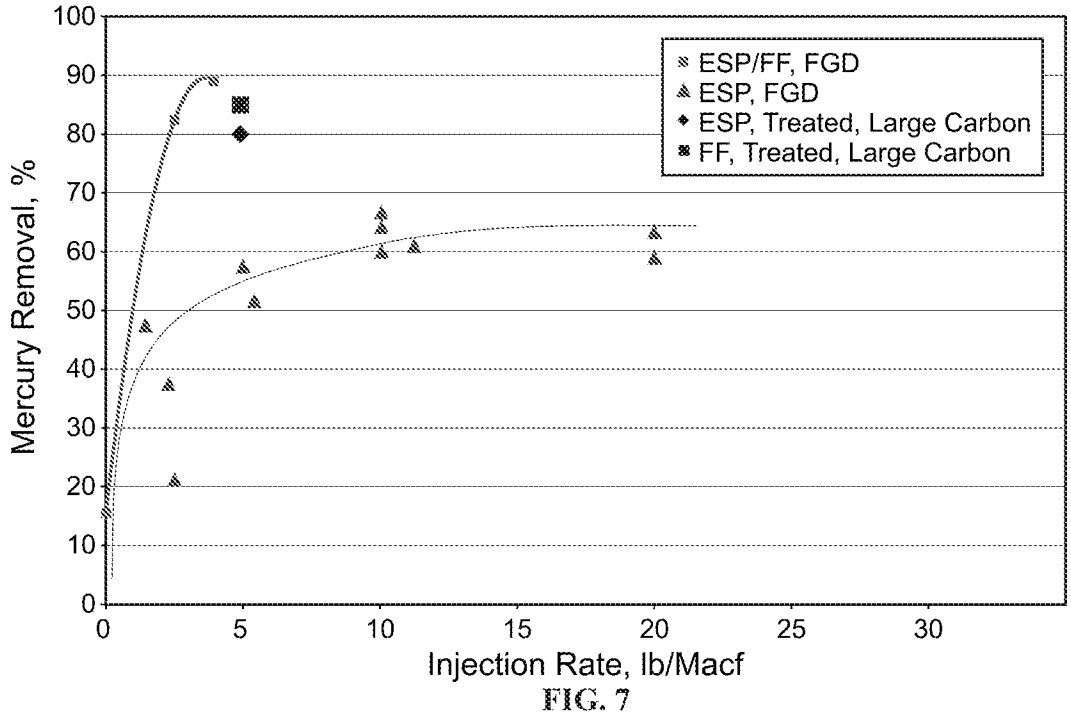

Mercury Removal, %
100
90
80
70
60
50
40
30
20
10
0
0
5
10
15
20
25
30
Injection Rate, lb/Macf
ESP/FF, FGD
ESP, FGD
ESP, Treated, Large Carbon
FF, Treated, Large Carbon

FIG. 7

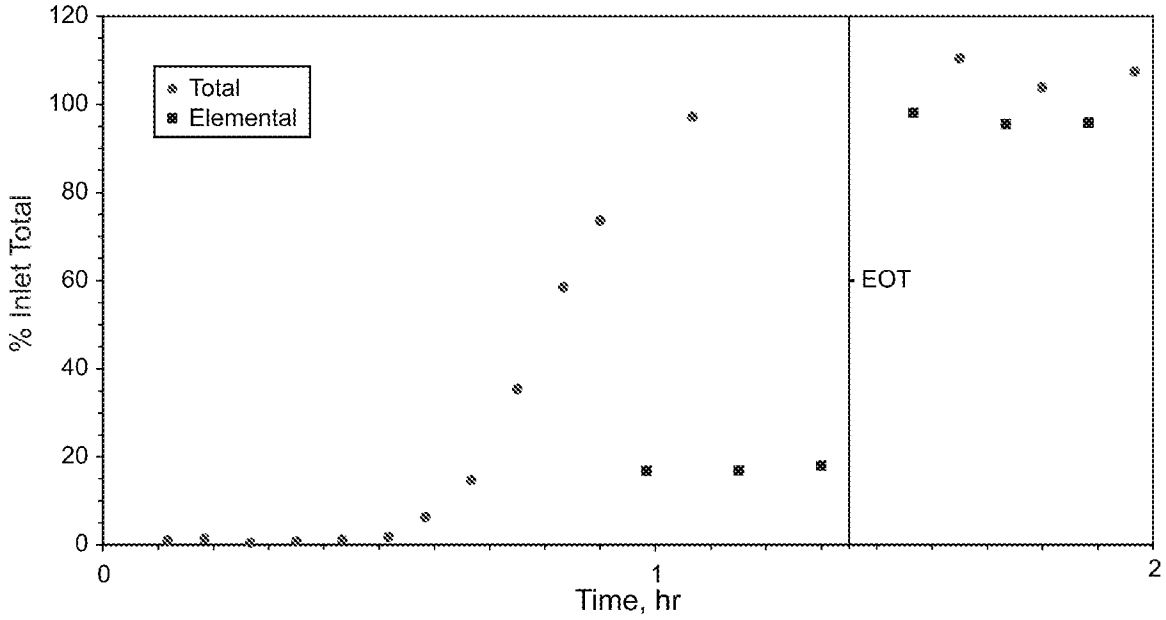

120
100
80
60
40
20
0
% Inlet Total
Total
Elemental
EOT
0
1
2
Time, hr

FIG. 8

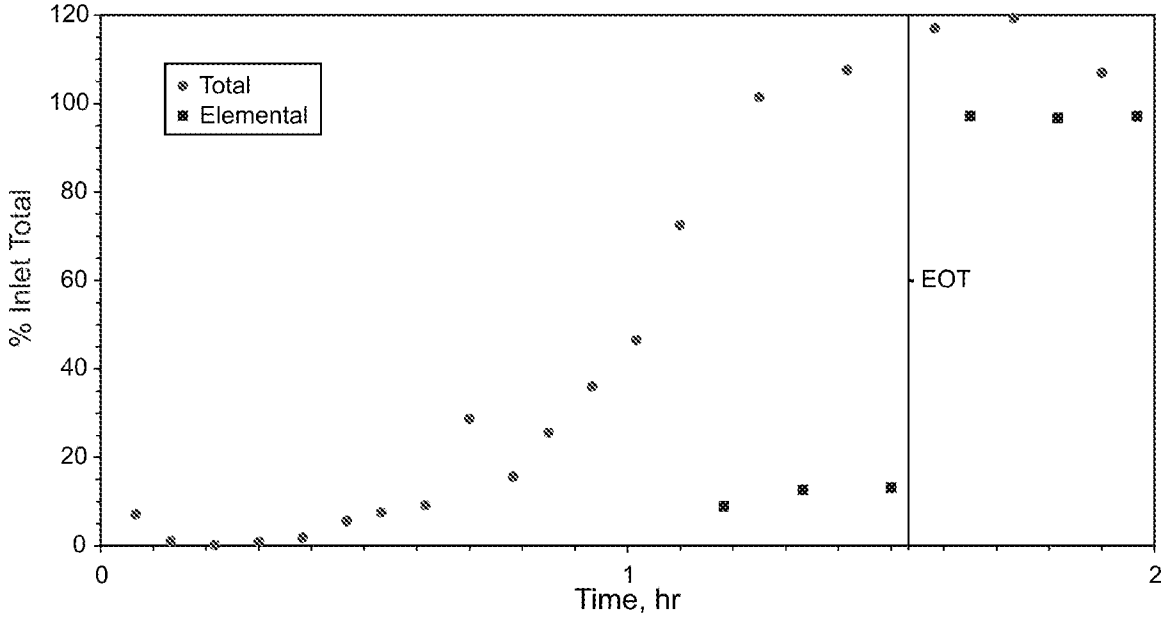

120
100
80
60
40
20
0
% Inlet Total
Total
Elemental
EOT
0
1
2
Time, hr

FIG. 9

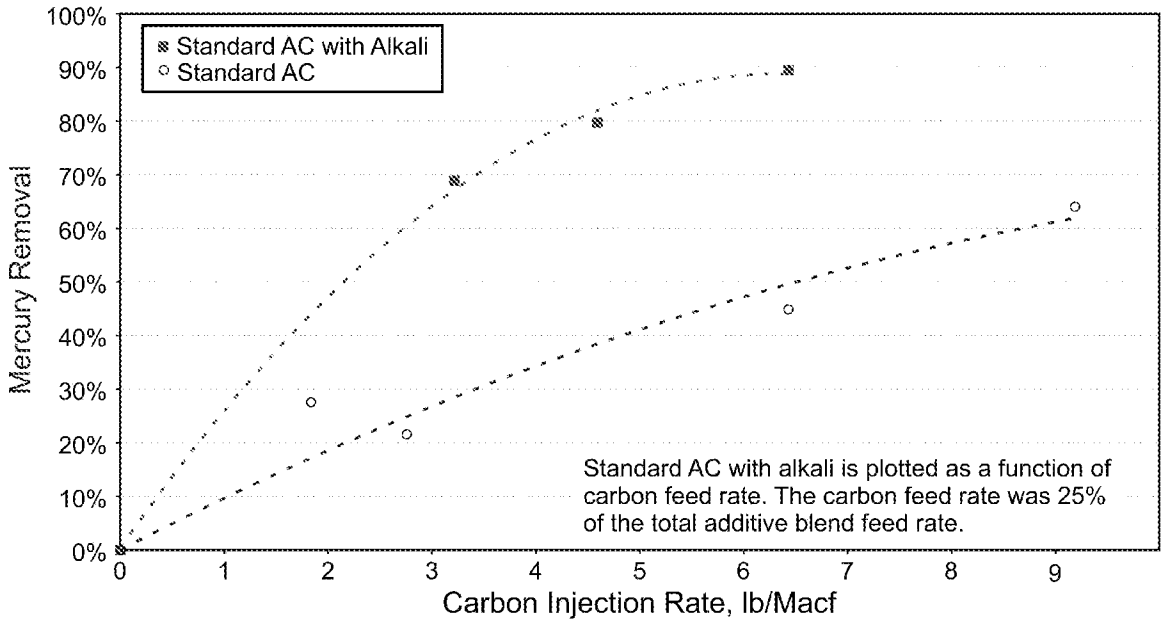

Standard AC with Alkali
Standard AC
Mercury Removal
100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
0
1
2
3
4
5
6
7
8
9
Carbon Injection Rate, lb/Macf
Standard AC with alkali is plotted as a function of carbon feed rate. The carbon feed rate was 25% of the total additive blend feed rate.

FIG. 10

Electronic Patent Application Fee Transmittal

Application Number:	12201595
Filing Date:	29-Aug-2008
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Filer:	Timothy S. Westby/Grace A. Wise
Attorney Docket Number:	2380-00207

Filed as Small Entity

Utility under 35 USC 111(a) Filing Fees

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Pages:				
Claims:				
Miscellaneous-Filing:				
Late filing fee for oath or declaration	2051	1	65	65
Petition:				
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
Extension-of-Time:				

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Miscellaneous:				
			Total in USD ($)	**65**

Electronic Acknowledgement Receipt

EFS ID:	4289738
Application Number:	12201595
International Application Number:	
Confirmation Number:	9735
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Customer Number:	23505
Filer:	Timothy S. Westby/Grace A. Wise
Filer Authorized By:	Timothy S. Westby
Attorney Docket Number:	2380-00207
Receipt Date:	14-NOV-2008
Filing Date:	29-AUG-2008
Time Stamp:	14:42:00
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	Deposit Account
Payment was successfully received in RAM	$65
RAM confirmation Number	11335
Deposit Account	032769
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Application Number: 12201595 Document Date: 11/14/2008

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HOUSTON, TX 77253-3267

FILING RECEIPT

OC000000032016576

Date Mailed: 09/17/2008

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

Applicant(s)

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Assignment For Published Patent Application

ENERGY & ENVIRONMENTAL RESEARCH CENTER FOUNDATION, Grand Forks, ND

Power of Attorney: None

Domestic Priority data as claimed by applicant

This application is a DIV of 11/209,163 08/22/2005
which claims benefit of 60/605,640 08/30/2004

Foreign Applications

If Required, Foreign Filing License Granted: 09/10/2008

The country code and number of your priority application, to be used for filing abroad under the Paris Convention, is **US 12/201,595**

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

Non-Publication Request: No

Early Publication Request: No
** **SMALL ENTITY** **

Title

SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

Preliminary Class

095

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Title 37, Code of Federal Regulations, 5.11 & 5.15

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APPLICATION NUMBER	FILING OR 371(C) DATE	FIRST NAMED APPLICANT	ATTY. DOCKET NO./TITLE
12/201,595	08/29/2008	Edwin S. Olson	2380-00207

CONFIRMATION NO. 9735

FORMALITIES LETTER

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OC00000032016577

Date Mailed: 09/17/2008

NOTICE TO FILE MISSING PARTS OF NONPROVISIONAL APPLICATION

FILED UNDER 37 CFR 1.53(b)

Filing Date Granted

Items Required To Avoid Abandonment:

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

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

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

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

- Replacement drawings in compliance with 37 CFR 1.84 and 37 CFR 1.121(d) are required. The drawings submitted are not acceptable because:
  - The drawings have a line quality that is too light to be reproduced (weight of all lines and letters must be heavy enough to permit adequate reproduction) or text that is illegible (reference characters, sheet numbers, and view numbers must be plain and legible) see 37 CFR 1.84(l) and (p)(1)); See Figure(s) 2, 4-10.
  - Numbers, letters, and reference characters on the drawings must measure at least 0.32 cm (1/8 inch) in height. See Figure(s) 4-10.

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

The applicant needs to satisfy supplemental fees problems indicated below.

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- To avoid abandonment, a surcharge (for late submission of filing fee, search fee, examination fee or oath or declaration) as set forth in 37 CFR 1.16(f) of $**65** for a small entity in compliance with 37 CFR 1.27, must be submitted with the missing items identified in this notice.

SUMMARY OF FEES DUE:

Total additional fee(s) required for this application is $**65** for a small entity
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SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

CROSS-REFERENCE TO RELATED APPLICATIONS

[0001] This application is a divisional of U.S. Patent Application No. 11/209,163 filed August 22, 2005, entitled "Sorbents for the Oxidation and Removal of Mercury", which claims the benefit of U.S. Provisional Application Serial No. 60/605,640, filed August 30, 2004, both applications are hereby incorporated herein by reference in their entirety for all purposes.

STATEMENT REGARDING F EDERALLY SPONSORED RESEARCH OR DEVELOPMENT

[0002] This invention was made with United States Government support under Grant Numbers R 827649-01 and CR 830929-01 awarded by the United States Environmental Protection Agency and under Contract Number DE-FC26-98FT40320 awarded by the United States Department of Energy. The United States Government has certain rights in this invention.

BACKGROUND OF THE INVENTION

Technical Field of the Invention

[0003] 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

[0004] 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

[0005] 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.

[0006] 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.

[0007] 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

[0008] 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.

[0009] 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.

[0010] 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, and combinations thereof, such that the reaction product is effective for the removal of mercury from a gas stream.

[0011] 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 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 activated carbon may have a mass mean particle diameter greater than about 40 micrometers.

[0012] 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.

[0013] 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.

[0014] 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.

[0015] 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.

[0016] 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 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.

[0017] 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.

[0018] 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.

[0019] 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 in a flue gas with substantially lower sorbent requirements. Through enhanced sorbent reactivity, mercury removal per gram of sorbent is increase, thereby decreasing the capital and operating costs by decreasing sorbent requirements.

[0020] 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.

[0021] 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.

[0022] 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.

[0023] 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.

[0024] 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.

[0025] 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.

[0026] 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-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.

[0027] 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.

[0028] 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

[0029] For a more detailed description of the preferred embodiments of the present invention, reference will now be made to the accompanying drawings.

[0030] Figure 1 schematically illustrates methods for preparation of promoted carbon sorbents in accordance with the present invention.

[0031] Figure 2 illustrates a proposed mechanistic model of the chemical reactions resulting in the oxidation and capture of mercury.

[0032] 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.

[0033] 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.

[0034] 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.

[0035] Figure 6 is a bar chart illustrating pilot-scale mercury removal results, including large-size sorbent results.

[0036] Figure 7 is a diagram illustrating the effects of sorbent size and injection rate on mercury removal for ESPs and fabric filters.

[0037] Figure 8 is a diagram illustrating the breakthrough curves for a brominated NORIT Darco FGD sorbent with inert sand.

[0038] Figure 9 is a diagram illustrating the breakthrough curves for brominated NORIT Darco FGD sorbent with a co-injected alkali material.

[0039] Figure 10 is a plot of mercury removal vs. carbon injection rate with and without co-injection of alkali material.

DETAILED DESCRIPTION

[0040] 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.

[0041] 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.

[0042] 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.

[0043] 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 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.

[0044] 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.

[0045] 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.

[0046] 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.

[0047] 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.

[0048] 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.

[0049] 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.

[0050] 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).

[0051] 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.

[0052] 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.

[0053] 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 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.

[0054] 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.

[0055] 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.

[0056] 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.

[0057] 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 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.

[0058] 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.

[0059] 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.

[0060] 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 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 ebulliated 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.

[0061] 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.

[0062] 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..

[0063] 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.

[0064] 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 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.

[0065] 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.

[0066] 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.

[0067] 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

[0068] 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.

[0069] 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

[0070] 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 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.

[0071] 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.

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 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

[0072] 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).

Optional Alkali Component

[0073] 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.

[0074] 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

[0075] 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

[0076] 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 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.

[0077] 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

[0078] 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 conjunction 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

[0079] 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 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

[0080] 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

[0081] 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.

[0082] 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

[0083] 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

[0084] 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 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 and 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 so that the excess of nitrogen is released after the bromine is transferred 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.

[0085] 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

[0086] 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.

[0087] 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 μg/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 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.

[0088] 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).

[0089] 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

[0090] 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

[0091] 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 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.

[0092] 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

[0093] 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

[0094] 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

[0095] 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 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

[0096] 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.

[0097] 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

[0098] 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.

[0099] 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.

[00100] 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

[00101] 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").

[00102] 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 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

[00103] 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

[00104] 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.

[00105] 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 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/Ebulliated Bed Preparation

[00106] 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 ebulliated 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

[00107] 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).

[00108] 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

[00109] 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.

[00110] 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 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

[00111] 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.

[00112] 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*

[00113] 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 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

[00114] 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

[00115] 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

[00116] 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.

[00117] 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

[00118] 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 (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.

[00119] 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

[00120] 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.

[00121] 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.

[00122] 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.

[00123] 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.

CLAIMS

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, wherein the sorbent is effective for the removal of mercury from a gas stream located downstream of a boiler, said removal of mercury yielding a cleaned gas stream, and wherein the composition of the promoted carbon sorbent is controlled by the mercury content of the cleaned 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 an activated carbon matrix in a fixed or a moving bed structure.

4. The promoted carbon sorbent of claim 3 wherein the structure is selected from the group consisting of moving fiber screens and monoliths.

5. The promoted carbon sorbent of claim 2 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 5 wherein the ratio of base activated carbon, the promoter, and the optional alkaline component is chosen for optimum economic removal of mercury from the gas stream.

7. The promoted carbon sorbent of claim 2 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.

8. The promoted carbon sorbent of claim 7 wherein the promoter is selected from the group consisting of Br_2, HBr, and combinations thereof.

9. The promoted carbon sorbent of claim 7 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.

10. The promoted carbon sorbent of claim 2 further comprising an optional secondary component comprising a halogen or a hydrohalide such that the reactivity and mercury capacity of the sorbent are enhanced.

11. The promoted carbon sorbent of claim 10 wherein the concentration of the optional secondary component on the finished sorbent is within the range of from about 1 wt-% to about 15 wt-% of the concentration of the promoter on the finished sorbent.

12. The promoted carbon sorbent of claim 10 wherein the optional secondary component is selected from the group consisting of Group V halides, Group VI halides, HI, HBr, HCl, and combinations thereof.

13. The promoted carbon sorbent of claim 12 wherein the optional secondary component is selected from the group consisting of I_2, HI, and combinations thereof.

14. The promoted carbon sorbent of claim 13 wherein the promoter is substantially in vapor form when combined with the base activated carbon.

15. The promoted carbon sorbent of claim 13 wherein the promoter is combined with an organic solvent prior to reaction with the base activated carbon.

16. The promoted carbon sorbent of claim 13 wherein the promoter and optional secondary component are combined with the base activated carbon substantially simultaneously.

17. A method of preparing a promoted carbon sorbent comprising providing an activated carbon; and forming a reaction product by 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 downstream of a boiler, thus yielding a cleaned gas stream, and wherein the composition of the sorbent is adjusted based upon the mercury content of the cleaned gas stream.

18. The method of claim 17 wherein the activated carbon has an average particle size greater than that of the components of the gas stream such that the promoted carbon sorbent produced therefrom can be substantially separated by physical means from the cleaned gas stream.

19. The method of claim 18 wherein the reaction product has an average particle size distribution greater than about 40 micrometers.

20. The method of claim 17 further comprising preparing the sorbent in-flight by reacting the activated carbon, the promoter, and an optional secondary component to enhance the reactivity and mercury capacity of the sorbent.

21. The method of claim 17 further comprising continuously monitoring the mercury content of the cleaned gas stream.

22. The promoted carbon sorbent prepared by the method of claim 21.

23. The method of claim 17 wherein the sorbent comprises an activated carbon matrix in a fixed or a moving bed structure.

24. The method of claim 23 wherein the structure is selected from the group consisting of moving fiber screens and monoliths.

25. The method of claim 17 wherein the ratio of activated carbon to promoter is adjusted for optimization of economic removal of mercury from the gas stream.

26. The method of claim 17 wherein the reaction product comprises from about 1 to about 30 grams promoter per 100 grams activated carbon.

27. The method of claim 26 wherein the promoter is in an organic solvent when contacting the activated carbon.

28. The method of claim 26 further comprising providing an optional alkaline component to improve the effectiveness of mercury removal.

29. The method of claim 26 wherein the promoter is in the gas phase when contacting the activated carbon.

30. The method of claim 29 wherein the promoter is selected from the group consisting of Br_2, a Group V bromide, a Group VI bromide, and combinations thereof.

31. The method of claim 29 further comprising reacting the 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.

32. The method of claim 31 further comprising providing an optional alkaline component to improve the effectiveness of mercury removal.

33. The method of claim 31 wherein the promoter and optional secondary component are contacted substantially simultaneously with the activated carbon.

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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Application Data Sheet 37 CFR 1.76	Attorney Docket Number	2380-00207
	Application Number	
Title of Invention	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY	

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

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Attorney Docket Number	2380-00207	**Small Entity Status Claimed**	☒
Application Type	Nonprovisional		
Subject Matter	Utility		
Suggested Class (if any)		**Sub Class (if any)**	
Suggested Technology Center (if any)			
Total Number of Drawing Sheets (if any)	7	**Suggested Figure for Publication (if any)**	1

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Application Data Sheet 37 CFR 1.76	Attorney Docket Number	2380-00207
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Title of Invention	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY	

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Prior Application Status	Pending		Remove
Application Number	Continuity Type	Prior Application Number	Filing Date (YYYY-MM-DD)
	Division of	11209163	2005-08-22
Prior Application Status	Expired		Remove
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11209163	non provisional of	60605640	2004-08-30

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Application Number	Country [i]	Parent Filing Date (YYYY-MM-DD)	Priority Claimed
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Title of Invention	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY	

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First Name	Timothy S.	Last Name	Westby	Registration Number	52352

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

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

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

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

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

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

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

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

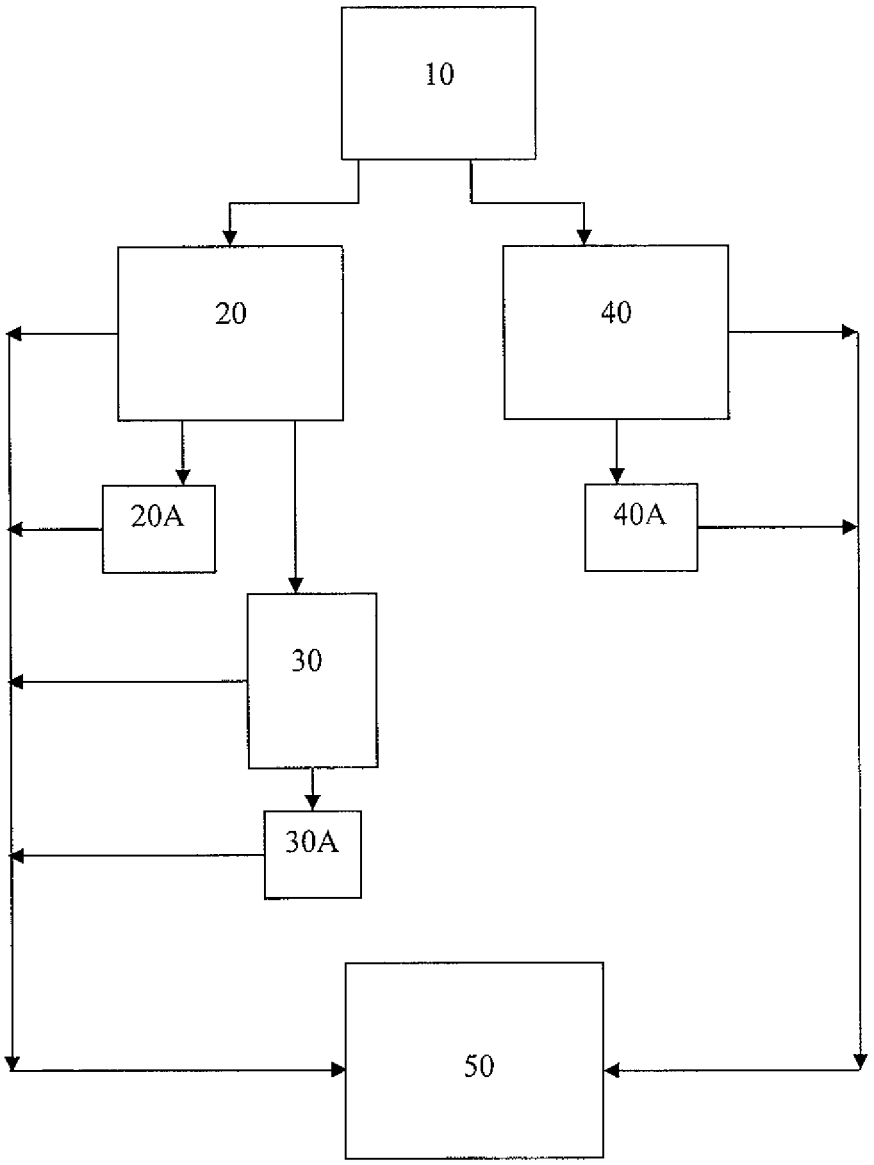


FIG. 1

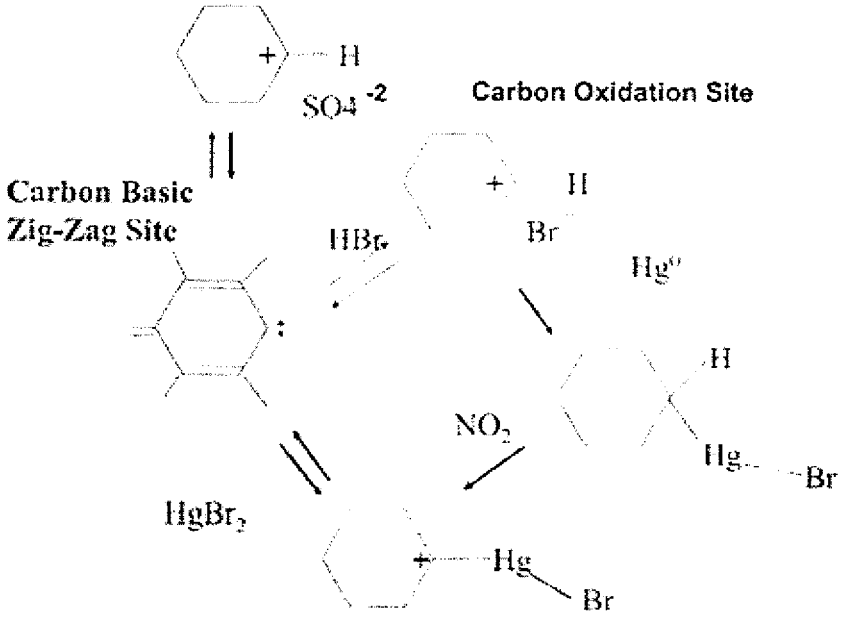

Carbon Basic
Zig-Zag Site
Carbon Oxidation Site
HBr
Hg°
NO2
HgBr2
Br
Hg
Br

FIG. 2

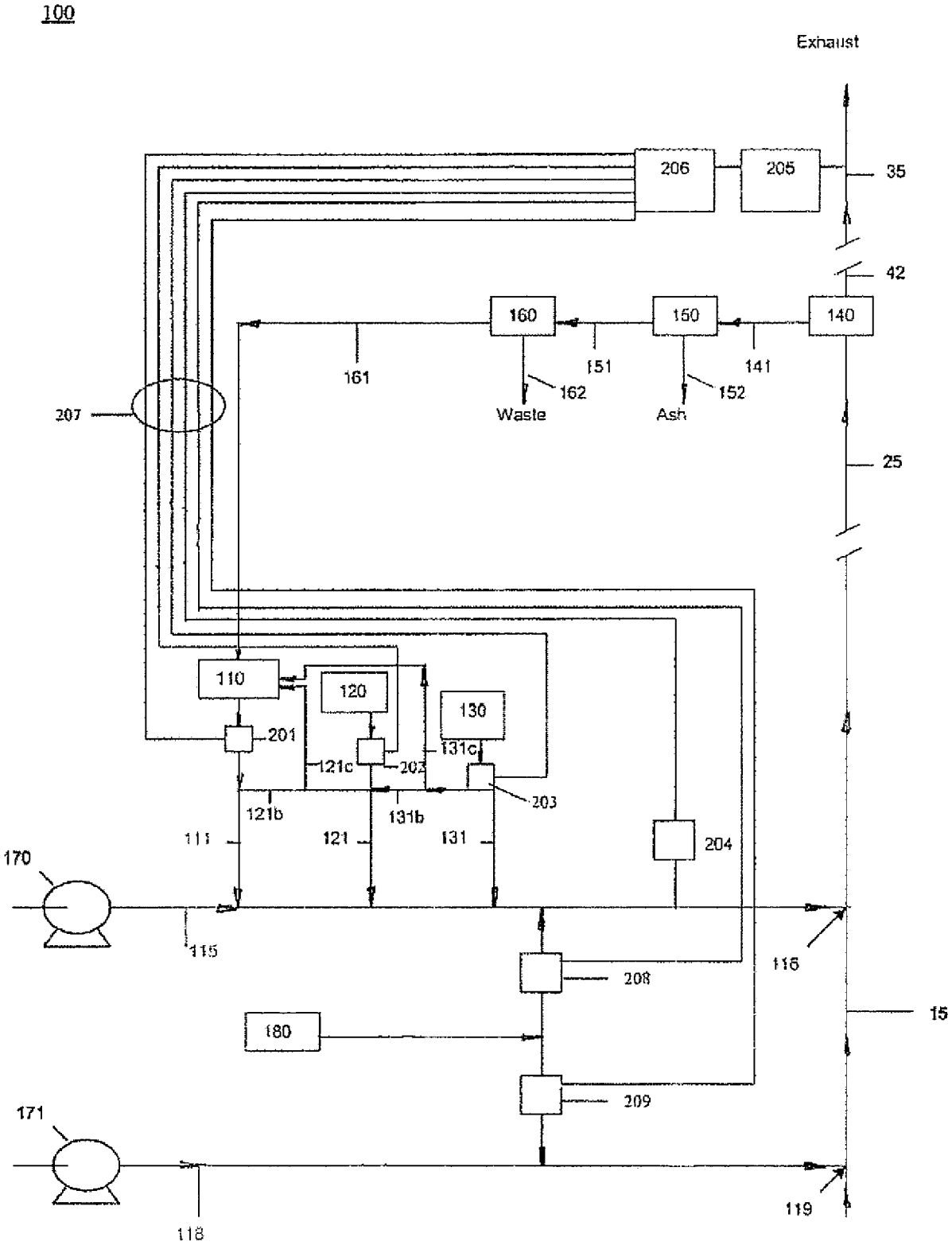

100
Exhaust
206
205
35
42
160
150
140
161
151
141
162
152
Waste
Ash
207
25
110
120
130
201
121c
202
131c
203
121b
131b
111
121
131
204
170
115
116
208
15
180
209
171
118
119

FIG. 3

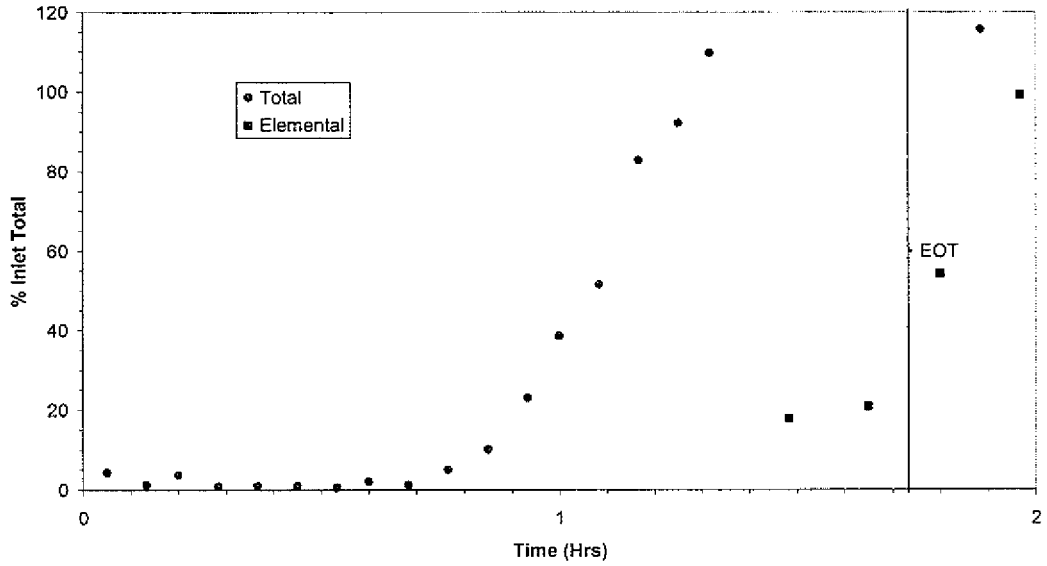

120
100
80
60
40
20
0
% Inlet Total
Total
Elemental
EOT
0
1
2
Time (Hrs)

FIG. 4

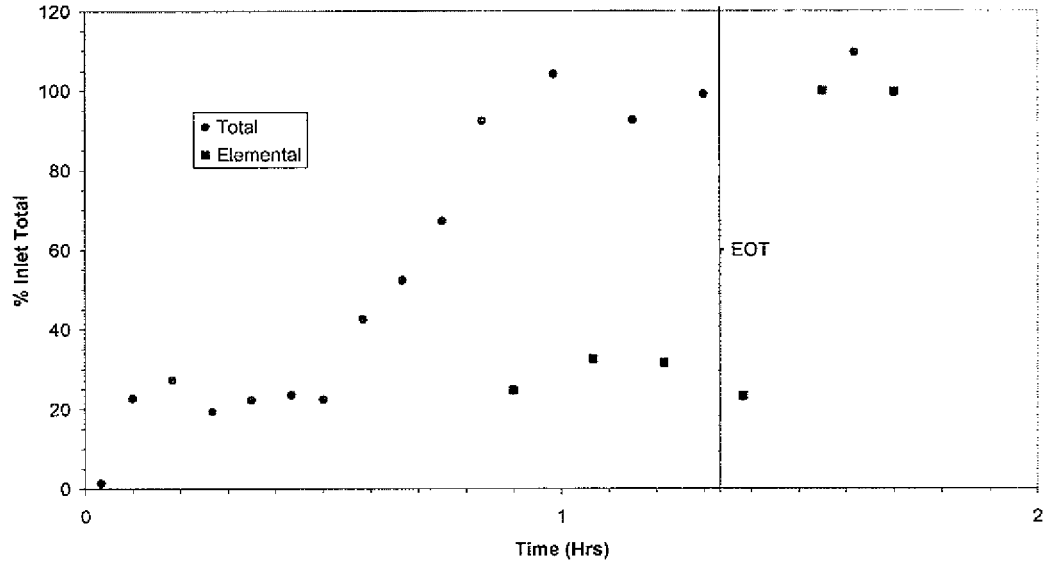

120
100
80
60
40
20
0
% Inlet Total
Total
Elemental
EOT
0
1
2
Time (Hrs)

FIG. 5

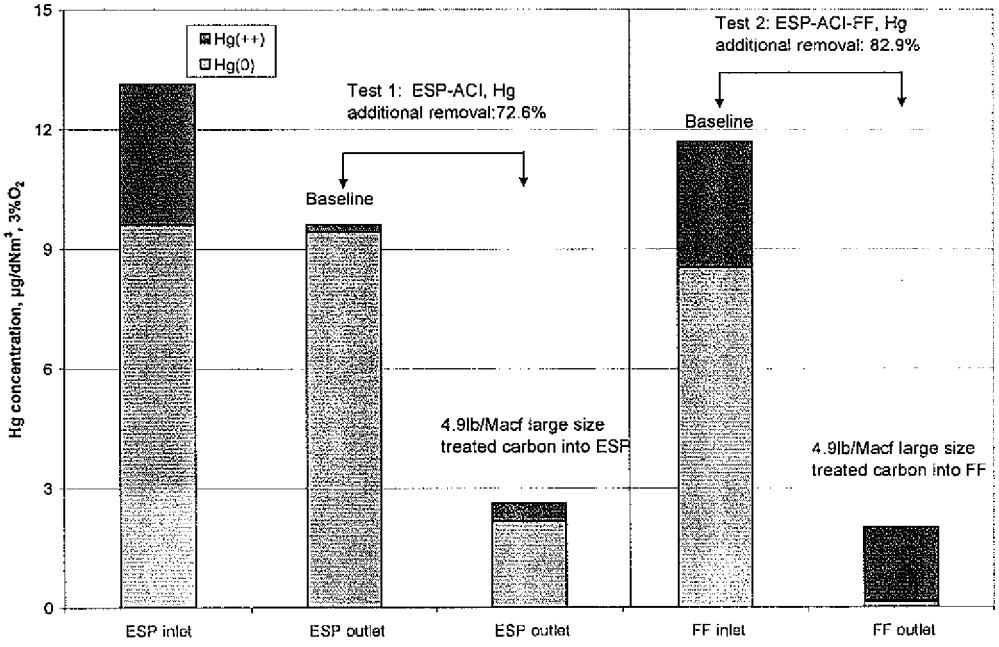

Hg(++)
Hg(0)
Test 1: ESP-ACI, Hg additional removal:72.6%
Baseline
4.9lb/Macf large size treated carbon into ESP
Test 2: ESP-ACI-FF, Hg additional removal: 82.9%
Baseline
4.9lb/Macf large size treated carbon into FF
Hg concentration, µg/dNm3, 3%O2
0
3
6
9
12
15
ESP inlet
ESP outlet
ESP outlet
FF inlet
FF outlet

FIG. 6

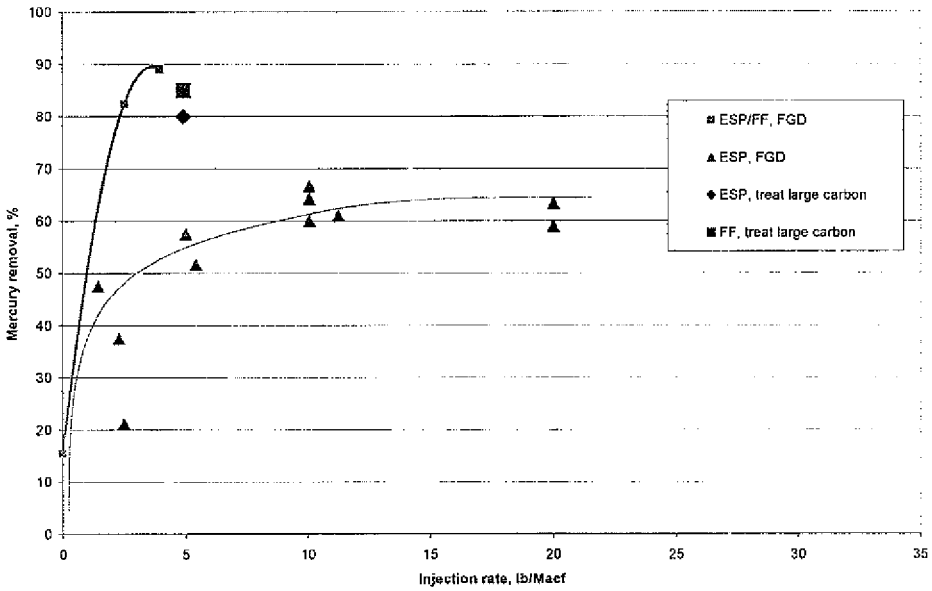

ESP/FF, FGD
ESP, FGD
ESP, treat large carbon
FF, treat large carbon
Mercury removal, %
Injection rate, lb/Macf
0
10
20
30
40
50
60
70
80
90
100
0
5
10
15
20
25
30
35

FIG. 7

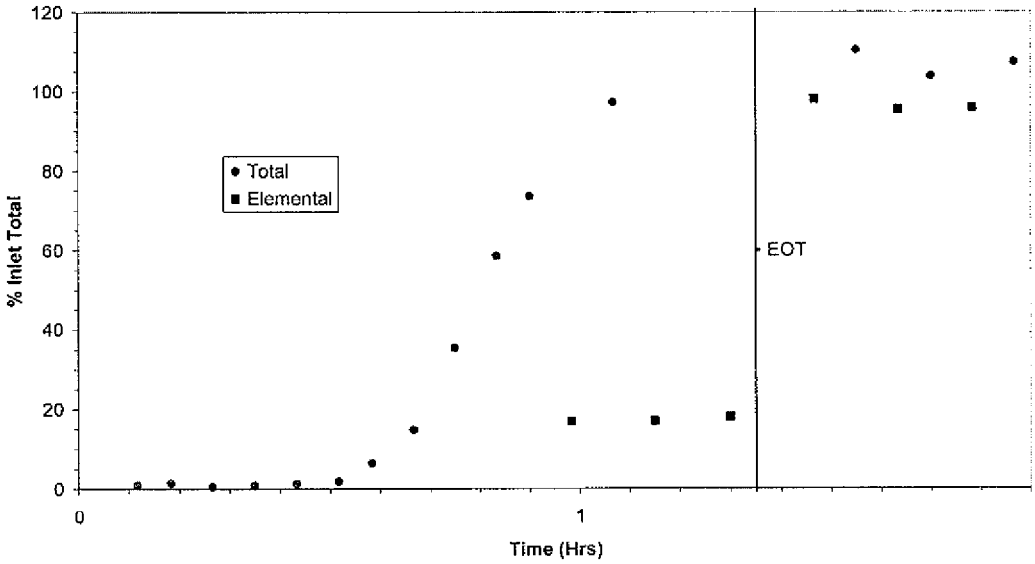

120
100
80
60
40
20
0
% Inlet Total
Total
Elemental
EOT
0
1
Time (Hrs)

FIG. 8

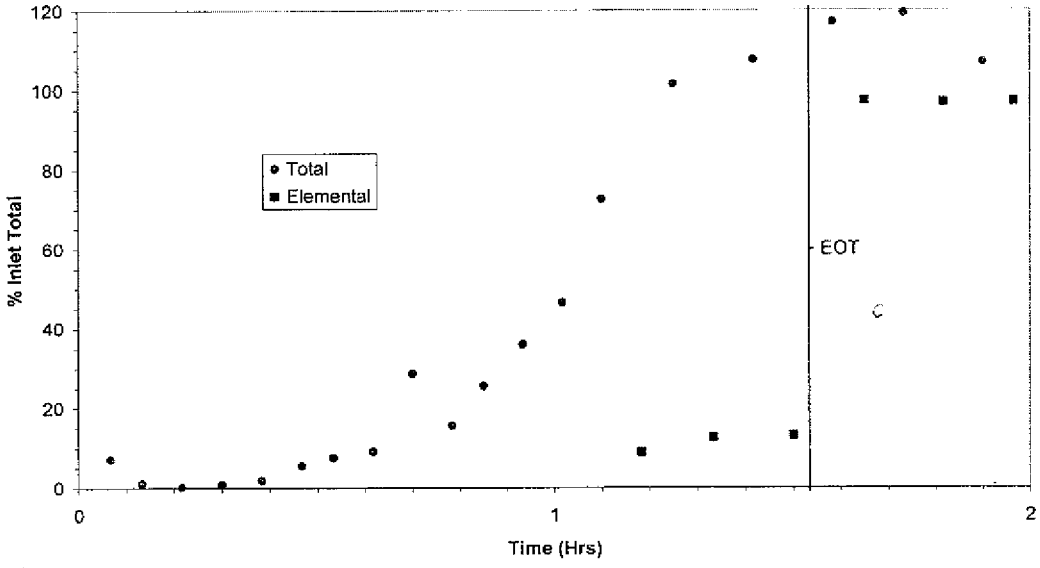

120
100
80
60
40
20
0
% Inlet Total
Total
Elemental
EOT
0
1
2
Time (Hrs)

FIG. 9

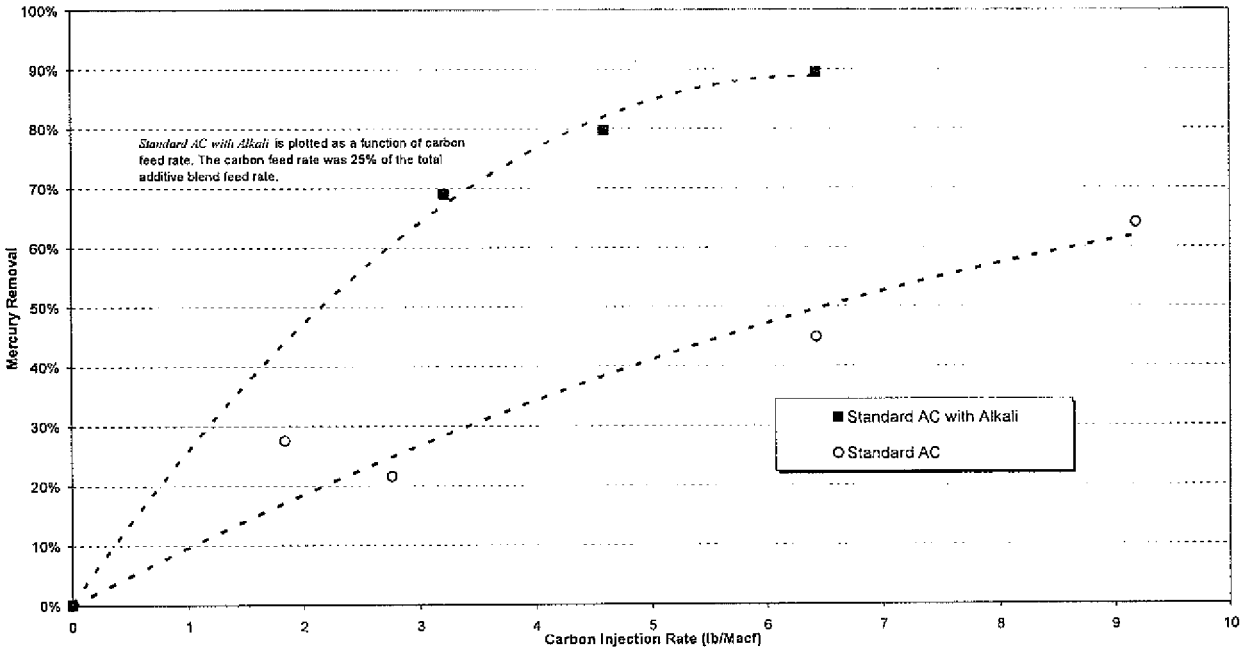

100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
Mercury Removal
Standard AC with Alkali is plotted as a function of carbon feed rate. The carbon feed rate was 25% of the total additive blend feed rate.
Standard AC with Alkali
Standard AC
0
1
2
3
4
5
6
7
8
9
10
Carbon Injection Rate (lb/Macf)

FIG. 10

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:	Timothy S. Westby/Grace A. Wise
Attorney Docket Number:	2380-00207

Filed as Small Entity

Utility under 35 USC 111(a) Filing Fees

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Basic Filing:				
Utility filing Fee (Electronic filing)	4011	1	75	75
Utility Search Fee	2111	1	255	255
Utility Examination Fee	2311	1	105	105
Pages:				
Claims:				
Claims in excess of 20	2202	13	25	325
Miscellaneous-Filing:				
Petition:				

Description	Fee Code	Quantity	Amount	Sub-Total in USD($)
Patent-Appeals-and-Interference:				
Post-Allowance-and-Post-Issuance:				
Extension-of-Time:				
Miscellaneous:				
		Total in USD ($)		**760**

Electronic Acknowledgement Receipt

EFS ID:	3865001
Application Number:	12201595
International Application Number:	
Confirmation Number:	9735
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Customer Number:	23505
Filer:	Timothy S. Westby/Grace A. Wise
Filer Authorized By:	Timothy S. Westby
Attorney Docket Number:	2380-00207
Receipt Date:	29-AUG-2008
Filing Date:	
Time Stamp:	15:54:44
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	Deposit Account
Payment was successfully received in RAM	$760
RAM confirmation Number	1226
Deposit Account	032769
Authorized User	
The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows: Charge any Additional Fees required under 37 C.F.R. Section 1.16 (National application filing, search, and examination fees)	

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1		DIVApplication.pdf	227519 c45773440b4e0ee1416a5046f113d6ea5f3897d0	yes	34
	Multipart Description/PDF files in .zip description				
	Document Description		**Start**	**End**	
	Specification		1	29	
	Claims		30	33	
	Abstract		34	34	
Warnings:					
Information:					
2	Application Data Sheet	ADS.pdf	1125366 d4d7904483be02b915f30be04be2d34667b88c95	no	5
Warnings:					
Information:					
3	Drawings-only black and white line drawings	FIGURES.pdf	87172 da20b87b837bd24f157febbbe578d75993b724fa	no	7
Warnings:					
Information:					
4	Fee Worksheet (PTO-06)	fee-info.pdf	36779 9a63a2463fde8ee6213d2873b581a2195055b3de	no	2
Warnings:					
Information:					
		Total Files Size (in bytes):	1476836		

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

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

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

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

New International Application Filed with the USPTO as a Receiving Office

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

Electronic Acknowledgement Receipt

EFS ID:	3865001
Application Number:	12201595
International Application Number:	
Confirmation Number:	9735
Title of Invention:	SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY
First Named Inventor/Applicant Name:	Edwin S. Olson
Customer Number:	23505
Filer:	Timothy S. Westby/Grace A. Wise
Filer Authorized By:	Timothy S. Westby
Attorney Docket Number:	2380-00207
Receipt Date:	29-AUG-2008
Filing Date:	
Time Stamp:	15:54:44
Application Type:	Utility under 35 USC 111(a)

Payment information:

Submitted with Payment	yes
Payment Type	Deposit Account
Payment was successfully received in RAM	$760
RAM confirmation Number	1226
Deposit Account	032769
Authorized User	
The Director of the USPTO is hereby authorized to charge indicated fees and credit any overpayment as follows: Charge any Additional Fees required under 37 C.F.R. Section 1.16 (National application filing, search, and examination fees)	

File Listing:

Document Number	Document Description	File Name	File Size(Bytes)/ Message Digest	Multi Part /.zip	Pages (if appl.)
1		DIVApplication.pdf	227519 c45773440b4e0ee1416a5046f113d6ea5f3897d0	yes	34
	Multipart Description/PDF files in .zip description				
	Document Description		**Start**	**End**	
	Specification		1	29	
	Claims		30	33	
	Abstract		34	34	
Warnings:					
Information:					
2	Application Data Sheet	ADS.pdf	1125366 d4d7904483be02b915f30be04be2d34667b88c95	no	5
Warnings:					
Information:					
3	Drawings-only black and white line drawings	FIGURES.pdf	87172 da20b87b837bd24f157febbbe578d75993b724fa	no	7
Warnings:					
Information:					
4	Fee Worksheet (PTO-06)	fee-info.pdf	36779 9a63a2463fde8ee6213d2873b581a2195055b3de	no	2
Warnings:					
Information:					
		Total Files Size (in bytes):	1476836		

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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.

08/29/08

Approved for use through 7/31/2006. OMB 0651-0032
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

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PATENT APPLICATION FEE DETERMINATION RECORD

Substitute for Form PTO-875

Application or Docket Number: **12/201595**

APPLICATION AS FILED – PART I

FOR	(Column 1) NUMBER FILED	(Column 2) NUMBER EXTRA	SMALL ENTITY RATE ($)	SMALL ENTITY FEE ($)	OR	OTHER THAN SMALL ENTITY RATE ($)	OTHER THAN SMALL ENTITY FEE ($)
BASIC FEE (37 CFR 1.16(a), (b), or (c))	N/A	N/A	N/A	75		N/A	
SEARCH FEE (37 CFR 1.16(k), (i), or (m))	N/A	N/A	N/A	255		N/A	
EXAMINATION FEE (37 CFR 1.16(o), (p), or (q))	N/A	N/A	N/A	105		N/A	
TOTAL CLAIMS (37 CFR 1.16(i))	33 minus 20 =	* 13	X 25=	325	OR	X 50=	
INDEPENDENT CLAIMS (37 CFR 1.16(h))	2 minus 3 =	*	X 105=			X 210=	
APPLICATION SIZE FEE (37 CFR 1.16(s))	If the specification and drawings exceed 100 sheets of paper, the application size fee due is $250 ($125 for small entity) for each additional 50 sheets or fraction thereof. See 35 U.S.C. 41(a)(1)(G) and 37 CFR						
MULTIPLE DEPENDENT CLAIM PRESENT (37 CFR 1.16(j))			N/A			N/A	
			TOTAL	760		TOTAL	

* If the difference in column 1 is less than zero, enter "0" in column 2.

APPLICATION AS AMENDED – PART II

AMENDMENT A		(Column 1) CLAIMS REMAINING AFTER AMENDMENT		(Column 2) HIGHEST NUMBER PREVIOUSLY PAID FOR	(Column 3) PRESENT EXTRA	SMALL ENTITY RATE ($)	SMALL ENTITY ADDITIONAL FEE ($)	OR	OTHER THAN SMALL ENTITY RATE ($)	OTHER THAN SMALL ENTITY ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	*	Minus	**	=	X =		OR	X =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=	X =		OR	X =	
	Application Size Fee (37 CFR 1.16(s))									
	FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))					N/A		OR	N/A	
						TOTAL ADD'T FEE		OR	TOTAL ADD'T FEE	

AMENDMENT B		(Column 1) CLAIMS REMAINING AFTER AMENDMENT		(Column 2) HIGHEST NUMBER PREVIOUSLY PAID FOR	(Column 3) PRESENT EXTRA	SMALL ENTITY RATE ($)	SMALL ENTITY ADDITIONAL FEE ($)	OR	OTHER THAN SMALL ENTITY RATE ($)	OTHER THAN SMALL ENTITY ADDITIONAL FEE ($)
	Total (37 CFR 1.16(i))	*	Minus	**	=	X =		OR	X =	
	Independent (37 CFR 1.16(h))	*	Minus	***	=	X =		OR	X =	
	Application Size Fee (37 CFR 1.16(s))									
	FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM (37 CFR 1.16(j))					N/A		OR	N/A	
						TOTAL ADD'T FEE		OR	TOTAL ADD'T FEE	

* If the entry in column 1 is less than the entry in column 2, write "0" in column 3.
** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 20, enter "20".
*** If the "Highest Number Previously Paid For" IN THIS SPACE is less than 3, enter "3".
The "Highest Number Previously Paid For" (Total or Independent) is the highest number found in the appropriate box in column 1.

This collection of information is required by 37 CFR 1.16. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.