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22764 U.S. PTO 083004

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INVENTOR(S)

Given Name (first and middle [if any])	Family Name or Surname	Residence (City and either State or Foreign Country)
Edwin S.	Olson	Grand Forks, ND USA
Michael J.	Holmes	Thompson, ND USA

Additional inventors are being named on the 1 separately numbered sheets attached hereto

TITLE OF THE INVENTION (500 characters max)

SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

Direct all correspondence to: CORRESPONDENCE ADDRESS:

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ENCLOSED APPLICATION PARTS (check all that apply)

☑ Specification Number of Pages 17
☐ CD(s), Number
☐ Drawing(s) Number of Sheets (included with spec.
☐ Other (specify)
☐ Application Data Sheet. See 37 CFR 1.76

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[Page 1 of 2]

Respectfully submitted,

SIGNATURE

Date 8-30-04

REGISTRATION NO. 37361 (if appropriate)

TYPED or PRINTED NAME Bruce A. Johnson

Docket Number: EER.P0023P

TELEPHONE 512-301-9900

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PROVISIONAL APPLICATION FOR PATENT COVER SHEET
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INVENTOR(S)/APPLICANT(S)		
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Number 1 of 1

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22764 U.S.PTO
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FEE TRANSMITTAL for FY 2001

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Complete if Known	
Application Number	TBD
Filing Date	
First Named Inventor	Edwin S. Olson
Examiner Name	
Group Art Unit	
Attorney Docket No.	EER.P0023P

TOTAL AMOUNT OF PAYMENT $ 80.00

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FEE CALCULATION

1. BASIC FILING FEE

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101	770	201	385	Utility filing fee	
106	340	206	170	Design filing fee	
107	490	207	245	Plant filing fee	
108	750	208	375	Reissue filing fee	
114	160	214	80	Provisional filing fee	$80

SUBTOTAL (1) $ 80.00

2. EXTRA CLAIM FEES

		Extra Claims		Fee from below		Fee Paid
Total Claims	-20**=	0	X	$9	=	$0.00
Independent Claims	-3**=	0	X	$43	=	$0.00
Multiple Dependent					=	

Large Fee Code	Entity Fee ($)	Small Fee Code	Entity Fee ($)	Fee Description
103	18	203	9	Claims in excess of 20
102	86	202	43	Independent claims in excess of 3
104	270	204	135	Multiple dependent claim, if not paid
109	80	209	40	** Reissue independent claims over original patent
110	18	210	9	** Reissue claims in excess of 20 and over original patent

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FEE CALCULATION (continued)

3. ADDITIONAL FEES

Large Fee Code	Entity Fee ($)	Small Fee Code	Entity Fee ($)	Fee Description	Fee Paid
105	130	205	65	Surcharge - late filing fee or oath	$0
127	50	227	25	Surcharge - late provisional filing fee or cover sheet	$0
139	130	139	130	Non-English specification	$0
147	2,520	147	2,520	For filing a request for *ex parte* reexamination	$0
112	920*	112	920*	Requesting publication of SIR prior to Examiner action	$0
113	1,840*	113	1,840*	Requesting publication of SIR after Examiner action	$0
115	110	215	55	Extension for reply within first month	$0
116	410	216	205	Extension for reply within second month	$0
117	890	217	446	Extension for reply within third month	$0
118	1.390	218	695	Extension for reply within fourth month	$0
128	1.890	228	945	Extension for reply within fifth month	$0
119	310	219	155	Notice of Appeal	$0
120	310	220	155	Filing a brief in support of an appeal	$0
121	270	221	135	Request for oral hearing	$0
138	1,510	138	1,510	Petition to institute a public use proceeding	$0
140	110	240	55	Petition to revive – unavoidable	$0
141	1,240	241	620	Petition to revive – unintentional	$0
142	1,240	242	620	Utility issue fee (or reissue)	$0
143	440	243	220	Design issue fee	$0
144	600	244	300	Plant issue fee	$0
122	130	122	130	Petitions to the Commissioner	$0
123	50	123	50	Petitions related to provisional applications	$0
126	240	126	240	Submission of Information Disclosure Stmt	$0
581	40	581	40	Recording each patent assignment per property (times number of properties)	$0
146	710	246	355	Filing a submission after final rejection (37 CFR § 1.129(a))	$0
149	710	249	355	For each additional invention to be examined (37 CFR § 1.129(b))	$0
179	710	279	355	Request for Continued Examination (RCE)	$0
169	900	169	900	Request for expedited examination of a design application	$0

Other fee (specify) _____ $0

* Reduced by Basic Filing Fee Paid **SUBTOTAL (3)** $0.00

SUBMITTED BY				Complete (if applicable)	
Name (Print/Type)	Bruce A. Johnson	Registration No. (Attorney/Agent)	37361	Telephone	512-301-9900
Signature				Date	8-30-04

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United States Provisional Patent Application for

SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

Inventors:
Edwin S. Olson
Michael J. Holmes
John H. Pavlish

ABSTRACT

A reactive sorbent material is described for capture of mercury in a vapor stream. The vapor stream includes flue gas from coal combustion or incineration, synthesis gas from gasification, as well as offgases from mineral processing, metal refining, retorting, cement manufacturing, chloralkali plants, dental facilities, and crematories. The sorbent is especially suited to, but not limited to, use in low-chlorine flue or exhaust gases where the mercury is mainly elemental. The sorbent comprises a bromide- modified carbon form containing a reactive compound produced by the reaction of bromine with the carbon. Other additional promoting substances can be added in smaller amounts to increase the reactivity with and sorption of elemental mercury. This combination results in a carbon bromide structure in the carbon lattice that can oxidize the mercury to a species that is strongly bonded to the carbon structure and, therefore, easily retained on the sorbent at higher temperatures in the presence of flue gas constituents. In some examples, the sorbent consists of a fine or coarse powdered material for injection into the gas stream or a granular or fibrous material for use in a fixed or moving bed. In some examples, the sorbent does not require in situ activation (an induction period) in a flue gas stream as do conventional activated carbon sorbents. Other additives of base chemistry (Ca, Na, and others) may be added to flue gas as a sorbent mixture, or co-injected to selectively sorb acid gases or other flue gas constituents to keep available reactive sites for mercury oxidation and subsequent capture. The sorbent can be regenerated by washing off contaminating components derived from flue gas that poison the spent sorbent. Novel methods for introduction of the sorbent dispersion into the mercury contaminated gas stream are described.

OUTLINE OF EXAMPLES OF THE INVENTION

Following is an outline of examples of the invention. The invention may also take on various other examples or embodiments, as a person skilled in the art would understand.

1. A modified carbon sorbent was prepared by reacting the carbon with molecular bromine by applying a solution of molecular bromine in an organic solvent to the carbon, followed by removal of the modified carbon from the bulk of solvent and further removal of solvent from the pores by drying the carbon in air or nitrogen. The solution may also contain an additional promoting substance in small amounts. In one example, the amount of bromine taken up by the carbon ranges from <1 to about 30 g per 100g of activated carbon, depending on the proportions used.

1A. (Dependent)
Same as 1, carbon is powdered activated carbon.
Carbon is granular activated carbon.
Carbon is very fine carbon black.
Carbon is very fine carbon fiber.
Carbon is aerogel carbon film.
Carbon is char recovered from pyrolysis process.
Carbon is powdered activated carbon of size distribution larger than fly ash size distribution, so that it can be physically separated from the ash and regenerated for reuse.
Carbon is regenerated from previous usage cycle and recycled .
Other desired types of carbon may also be used.

1B. (Dependent)
Same as 1, Solvent is chlorinated hydrocarbon such as dichloromethane
Same as 1, Solvent is hydrocarbon solvent such as petroleum ether or ligroin or pentane or hexane or toluene or benzene.
Same as 1, Solvent is water.
Same as 1, Solvent is carbon disulfide.
Same as 1, Solvent is a waste solvent.
Same as 1, Solvent is an ether.
Same as 1, Solvent is recycled from previous batch.
Same as 1, Solvent is supercritical solvent such as CO_2.
Other desired types of Solvents may also be used.

1C. (Dependent)
Same as 1, solution of bromine is applied by stirring the carbon in the bromine solution.
Same as 1, solution of bromine is applied by spraying a solution of molecular bromine in an organic solvent, followed by removal of solvent from the pores by drying the carbon in air or nitrogen.
The bromine may also be applied using other desired techniques.

1D. (Dependent)
Same as 1, removal of solvent is by filtration, centrifugation, settling, and decantation and drying in vacuum, air, or other gas.
Same as 1, when sprayed or minimal volume, removal of solvent is by drying in air, vacuum, or other gas.
The solvent may also be removed using other desired techniques.

1E. (Dependent)
Same as 1, additional substance is iodine, at 1%B10% of the bromine concentration added.
Same as 1, additional substance is hydrohalide (HI, HBr, HCl), etc., 1%B-10 % of bromine.
Same as 1, additional substance is a compound comprised of a Group V or VI element or combination of Group V or VI with Group VII element, such as SCl_2, 1-10 wt % of bromine.

2. A modified carbon sorbent was prepared by reacting the carbon with molecular bromine by contacting the carbon with a gas containing molecular bromine. In one example, the amount of bromine taken up by the carbon ranges from <1 to about 30 g per 100g of activated carbon, depending on the proportions used.

Contacting is by passing the Br_2 gas through a bed of the carbon.
Bed is ebullated or fluidized.
Bed is moving bed of granules or fibers.
Contacting is by admitting Br_2 gas to a duct through which the carbon is transported from a reservoir to the flue gas duct via an air-driven entrainment method, thus promoting the activity of the carbon in-flight.

Same additives as 1E, except these are added in the gas phase, and added in-flight as in 2 above.

3. A modified activated carbon sorbent was prepared by reacting the carbon with hydrogen bromide (HBr) by stirring the carbon in a solution of HBr in an organic solvent or in water, followed by removal of the modified carbon from the bulk of solvent and further removal of solvent from the pores by drying the carbon in air or nitrogen. In one example, the amount of bromine taken up by the carbon ranges from <1 to about 30 g per 100g of activated carbon, depending on the proportions used.

Same dependent clauses as in 1A with regard to carbon forms. Same solutions and treatment as 1B,1C, 1D.

Same additives as 1E, added into the solution phase or later in the gas phase, and added in-flight as in 2 above.

4. An activated carbon sorbent was prepared by impregnating hydrogen bromide by passing a gas composition containing hydrogen bromide diluted with nitrogen or air through a fluidized bed or ebullated bed of the carbon. In one example, the amount of bromine taken up by the carbon ranges from <1 to about 30 g per 100g of activated carbon, depending on the proportions used.

Same dependent clauses as in 1A with regard to carbon forms.
Contacting options are similar to 2.

5. Sequential treatment of activated carbon with bromine or HBr, followed by another mercury stabilizing reagent to generate a highly reactive bifunctional sorbent, the reagent to include sulfur and selenium species, such as H_2S, SO_2, H_2Se, Se, SeO_2, CS_2, P_2S_5.

6. Sequential treatment of activated carbon with mercury stabilizing reagent, followed by bromine or HBr to generate a highly reactive bifunctional sorbent, the reagent to include sulfur and selenium species, such as H_2S, SO_2, H_2Se, Se, SeO_2, CS_2, P_2S_5.

7. Treatment of a carbon with a Group V or VI element combined with Group VII element, such as PBr_3 or SCl_2. In one example, the amount of Group V or VI halide is 1-30 wt % of the carbon. The compound is added in vapor phase or in solution (solvent is hydrocarbon, chlorinated hydrocarbon, or other solvent as described in 1B, but not water, since the compound is destroyed in water solution).

8. The process for preparing the bromine-containing activated carbon sorbent as described in Examples 1-6 and coupled with the injection of the bromine-containing fine-particle sorbent into the contaminated gas stream and subsequent removal of the fine particles of sorbent from the gas stream in a collection device, such as bag filtration of electrostatic precipitator (ESP). .

9. The process of using additives (1-8) in conjunction with sorbents to capture acid gases and other flue gas constituents that block oxidation reactions or consume sorbent sites that would otherwise capture mercury. The additive is a compound comprised of Group I or II elements, such as Ca, Na, and others. These additives of base chemistry may be added to flue gas as a sorbent mixture or co-injected to selectively sorb acid gases or other gas constituents that compete for mercury reactive/sorption sites, thereby improving the effectiveness of activated carbon to capture mercury. In addition, the base additives can augment the treated activated carbon by helping to capture oxidized mercury forms in the flue gas, such as mercuric chloride, or that are produced on the carbon sorbent, such as mercuric bromide, but are released to the gas phase as the sorbent becomes saturated or capacity limited.

10. The process of preparing a larger-than-normal (>40 μm) sorbent particle size, promoting with the halogen treatment as described in 1-8, using the treated sorbent for capture of mercury by injecting in the flue gas, and finally separating and recovering the sorbent from the ash particles. The larger sorbent size thus will allow easy mechanical/physical separation so that sorbent can be regenerated, recycled, and reused, and the ash can be sold as a low-carbon cementatious byproduct for concrete. The separation can be effected by various methods, such as density (gravity), floatation, or sieving methods.

11. The process for preparing the bromine-containing activated carbon sorbent as described in Example 1-10, coupled with the direct injection of the slurry of the prepared fine-particle sorbent in the solvent or aqueous phase, so that the aerosol particles of the slurry achieve a high degree of dispersion in the gas stream and evaporation of the solvent in the hot gas occurs rapidly. The amount of slurry added is flow-controlled, and the concentration of particles in the slurry is monitored by optical density measurements prior to injection. Collection of particles is performed.

12. The process for preparing the bromine-containing activated carbon sorbent, as described in Examples 2 and 4, coupled where the fine-particle dispersion of brominated sorbent in the gas stream at the top of the ebullated bed is led to the flue gas or offgas duct where the said dispersion is contacted with mercury-contaminated flue gas or offgas stream. The amount of aerosol dispersion added is controlled by the gas flow to the ebullated bed, and the concentration of particles in the dispersion can be monitored by optical density measurements prior to injection. Collection of particles is performed by the existing particulate-collection equipment.

13. The process for preparing the bromine-containing activated carbon sorbent bed, coupled with contacting the bed with the mercury-contaminated flue gas or offgas.

14. The process for preparing the bromine-containing activated carbonBsorbent, as described in Examples 2 and 4, or untreated activated carbon, coated on a substrate or as activated carbon material. This material is placed in a moving system that geometrically allows for control of rate of exposure, sorption, and exchange of material in a continuous or semicontinuous manner.

15. In the sorbent and mercury removal process outlined in paragraphs above, the bromine reagents promote metal oxidation activity by creating cationic or radical character on the carbon edge structure that attracts mercury atoms and extracts electrons from said mercury atoms, resulting in oxidized mercury, which is more easily captured on a sorbent. A similar mechanism applies to oxidation and capture of reduced arsenic and other pollutant species.

DESCRIPTION OF THE INVENTION

Field of the Invention

The invention relates to the process for the removal of mercury and other pollutant species from gas streams generated during the burning of fossil fuels, such as in a coal-fired utility or synthesis gas from gasification facilities. This removal process would utilize a sorbent and/or a combination of additives to remove pollutants from the gas stream, which are at the same time interacting with other gas components that result in poisoning or deactivating the sorbent and reducing its capacity or reaction rates with the targeted pollutant species. The present invention provides a cost-effective way to capture the pollutants by utilizing exceptionally active carbon sorbents and promoting mercury sorbent surface reactions, containing a bromide form of the carbon, that capture mercury with high kinetic chemisorption (oxidation) activity, so that capture can occur during contact times of seconds or less. The sorbent does not require in situ activation (induction period) in the gas stream to achieve high activity as do conventional activated carbon sorbents. The reactivity of the sorbent toward the pollutants and the sorption capacity can be regenerated, recycled, and reused.

Background of the Invention

Burning fossil fuels generates a flue gas that contains mercury (and other trace elements) that was in the coal, plus oxides of sulfur and nitrogen (acid gas emissions) and particulates whose release to the environment must be controlled by use of sorbents, scrubbers, filters, precipitators, and other removal technologies. While control of other elements and/or flue gas pollutants may apply, the discussion provided is focused on mercury. Initially, mercury is released in the elemental form during combustion/gasification, but downstream of the boiler, in the ducts and stack of the combustion system, part of the elemental mercury is oxidized. The amount oxidized depends on the amount of acid gases present in the flue gas and other factors. Amounts of mercury vary with the coal, but a typical concentration of mercury in the stream of combustion gas is about 5 parts per billion (ppb). A pound of mercury or more per day may be emitted in some utilities.

Discussion of Sorbent Technologies

Several types of mercury control methods for flue gas have been investigated, including injection of fine sorbent particles into a flue gas duct and passing the flue gas through a sorbent bed. Fine-particle injection sorbents include activated carbon, metal oxide sorbent, sodium sulfide particles (for example, see U.S. Patent 6,214,304), and basic silicate or oxide sorbents (for example, see the Ghorishi reference (Ghorishi,B.; Gullet, B.K. Waste Manage Res. 1993, 16, 582). and Lancia references). When particle injection is employed, the mercury chemisorbed to the sorbent particle is removed from the gas stream in a bag house or ESP and collected along with ash particulates (for example, see U.S. Patents 4,889,698, 4,956,162, 5,672,323, 5,827,352, 6,027,551, and 5,505,766.). 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 approximately 60% was attained with a baghouse to collect carbon and ash. Even lower removal rates have been observed when using an ESP to collect the carbon, since the contact time of the carbon with the gas is very short.

A major problem with existing carbon injection systems is that the sorbent is initially unreactive, and only after exposure to the flue gas for about 20 minutes does the seasoned sorbent become effective and provide capture of 60% of the mercury in the gas. Consequently, these sorbents must be used in large amounts (high sorbent-to-mercury ratios) to effectively capture the mercury. These sorbents can be relatively expensive and cannot be easily separated from the ash, regenerated and reused. The collection of carbon in the ash also creates solid waste disposal problems, and the spent sorbent may contaminate the collected ash for use in various applications.

Use of Halogens in Mercury Capture

Methodologies for using halogens for the treatment of flue gas have not previously been effective. Halogen is defined as a member of the very active elements comprising Group VII-A of the periodic table. In the molecular elemental halogen form, including F_2, Cl_2 , Br_2, and I_2, the reaction with elemental mercury is not fast enough to result in the formation of oxidized mercury in a coal combustion flue gas with conventional ash control technology. The atomic elemental halogen form, which includes F atom, Cl atom, Br atom, and I atom, is about a million times more reactive to mercury, but the concentrations of these forms are extremely low, and also, therefore, cannot result in oxidation of mercury in a utility flue gas.

A halide is a compound formed from reaction of a halogen with another element or radical. In general, the halide compounds are normally very much less reactive than the molecular halogens, having expended their high chemical potential in forming the low energy halide. The halide is considered a reduced form and cannot therefore oxidize anything by itself. It is for this reason that it is not obvious that a halide-halogen treated activated carbon would be effective at oxidizing elemental mercury and provide effective capture of elemental mercury. In fact, for tests with salts, analytical data show the addition of bromine to an activated carbon or carbon black or soot does form a compound by reacting with the unsaturated groups on the carbon. By applying x-ray photoelectron spectroscopy, it has been determined that bromine or HBr added to carbon black forms chemical compounds within a carbon structure.

A body of theory has been developed from scientific evidence to explain the nature of the promotion effect of the bromine compounds. This principle is illustrated in FIG. 1, which in step 1 shows the reaction of the hydrogen bromide with the unsaturated structure of the activated carbon. This may be a carbene species on the edge of the graphene sheets 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.

We now teach that the formation of the new bromide compound with carbon increases the reactivity of the carbon forms toward mercury and other pollutants. The resulting bromide compound is uniquely suited to facilitate oxidation of the mercury. The effectiveness of the oxidation 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, which stabilizes the positive charge developing on the mercury and lowers 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 Br2 to the carbon forms a similar carbon bromide, in which the positive carbon oxidizes the mercury with the assistance of the bromide ion.

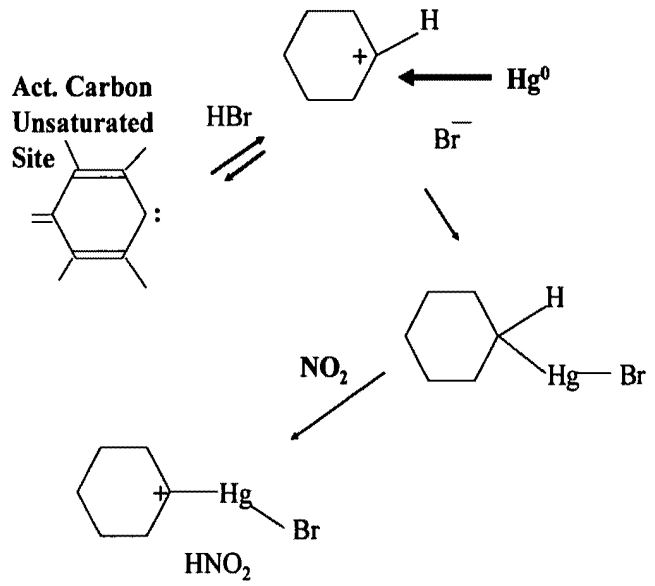

Figure 1. A mechanistic model of the reactions resulting in oxidation and capture of mercury.

Prior Art Publications

The earliest patents that relate to mercury capture on a sorbent are actually not for flue gas mercury control but for capture of mercury from air or other gas mixtures on a carbon or other sorbent. These patents were based on a model that recognized the affinity of halogens for mercury, rather than the formation of a reactive carbon. These patents are discussed below according to the type of halogen used for impregnation.

Iodine-Impregnated Sorbent

One patent that teaches a halogen impregnation (Stock, U.S. Patent 1,984,164, issued in 1934) describes the use of activated carbon or other solid impregnated with a halogen for removing Hg from air. The impregnation method is not specified. This seems to be the most general claim with respect to halogen impregnation. All the statements and claims appear to refer to halogen or iodine, but the patent does not appear to address bromides or other halides or bromide compounds or even specifically iodide or an iodine compound. The only example in the description is iodine impregnation. The patent also is restricted to air systems and did not pertain

to flue gas systems with reactive acid gases. The description did not assume that the iodine formed a reactive carbon species.

Japanese Patent JP 49-43197 describes the treatment of Hg-contaminated electrolysis cell gas using a metal iodide salt on a support. A similar treatment using a resin impregnated with a metal iodide is described in JP 50-6438. These patents do not appear to represent a carbon bromide compound as specified in this patent application.

Chlorine-Impregnated Sorbent

Another early patent (Hilgen, U.S. Patent 3,849,267, issued in 1974) describes a method for Hg removal from a gas which claims mixing molecular chlorine (Cl_2) with the gas to be cleaned and passing it through a nonporous sorbent such as glass wool and NaCl (not carbon). This was evidently not an obvious extension of the Stock patent, since a halogen was introduced as part of the gas phase being cleaned.

A later patent (deJong, et al., U.S. Patent 4,196,173, issued in 1980) describes a method for Hg removal in air which claims using a bed of activated carbon impregnated with halogen-only Cl_2 is specified in the other claims and examples, and it is prepared by flowing the Cl_2 stream through a carbon bed.

ICl-Impregnated Sorbent

An improved sorbent for mercury in air was patented by Revoir and Jones (U.S. Patent 3662523, issued in 1972), in which ICl and ICl_3 vapors were impregnated into the activated carbon. No explanation of the increased activity was provided. Since these compounds are interhalogen compounds, the patent did not infringe Stock.

HCl-Impregnated Sorbent

U.S. Patent 5,891,324 describes an activated carbon containing an acid (HCl, H_2SO_4, or H_3PO_4) for the removal of mercury contained in a liquid phase, such as would occur in a process steam in the oil industry. In this case, the model presented was that mercury is adsorbed from the liquid into the solid carbon phase at relatively low temperatures.

Bromine-Impregnated Sorbent (Nelson)

In U.S. Patent Application 2004/0003716, a method is described for removing mercury from a combustion gas. This method employs an exposure of a finely powdered activated carbon to a molecular bromine gas at an elevated temperature or to HBr to produce a sorbent with good mercury removal ability when injected into the flue gas duct. The model for the efficacy of the Nelson sorbent is not clearly stated, so it can be presumed that the reasoning for using bromine is the simple premise that some bromine form will help capture the mercury, as does iodine and chlorine. The methods for incorporation of the bromine in the Nelson application are not the same as with the present invention, and the Nelson method will not produce the identical carbon bromide species that are obtained in the art described in this application. The Nelson method lacks many of the features described in this application that impart exceptional activity to the sorbent in a convenient way, for example, the addition of smaller amount of a second more

powerful promoting agent, the use of facile solvent systems, including aqueous bromine, and the use of in-flight bromine treatment. Additionally, the patent application does not mention regeneration, recycling, or reuse, or the use of moving contactors or larger particle size to facilitate sorbent-ash separation.

SUMMARY OF THE INVENTION

A sorbent that is initially very reactive in oxidizing mercury can be used in very small amounts to achieve high capture efficiencies, thus lowering operation costs and lessening the disposal problem. In addition, further disposal reduction is made possible by recycling and reusing the sorbent that is produced using this technology. The time interval for the mercury and sorbent to interact in a flue gas duct and the subsequent collection of the mercury on the sorbent and ash may be very short- only seconds. This requires the sorbent to have high capacity and be very reactive toward mercury. A sorbent can be utilized in a very finely powdered form (e.g., 1-10 μm) to minimize mass transfer limitations. But, again, the reactivity should be very high to capture all of the mercury encountered by the fine particles. One feature of this invention is the process to prepare a sorbent containing a bromide 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 effective capture of the mercury.

One possible way to do this is to chemically combine molecular bromine from solution with activated carbon (edge sites). X-ray photoelectron spectroscopy established that the addition of bromine formed a chemical compound in the carbon structure. Thus, the sorbent does not represent a molecular bromine halogen form but a new chemically modified carbon structure. This may not occur with the less reactive iodine, where an I_2- molecular complex can exist on the carbon basal plane. In the bromine case, it is actually the carbon that oxidizes the mercury. Thus, an entirely new model is presented for the reactivity of the bromine-treated carbon with mercury. The reactive carbon form can be generated by the addition of bromine, hydrogen bromide, or combinations of bromine and other elements as described herein. Chlorine treatment resulted in lower-activity carbons because the chloride anion was less 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.

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, exceeding that of both the untreated carbon and the brominated carbon. The second compound comprises either a second halogen or a compound from a second halogen, such as HBr. 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).

Furthermore, we demonstrated that the sorbent can be readily treated with any combination of bromine and the second component in-flight using vapors of the these components contacting the

very fine carbon particles dispersed in air or other gas stream that conveys the particles to the flue gas duct. There is no particular temperature requirement for this contact. This technology is very simple to implement, so it will result in a great cost savings to the utility using this technology for mercury capture.

A unique, nonobvious technique for preparation of the treated carbon is through combining the treatment system with the carbon injection system at the end-use site. With this technique, the halogen is introduced to the carbon-air mixture in the transport line (or other part of the sorbent storage and injection system). Benefits over current concepts to treat sorbents off-site include the following:

- 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.
- This process uses existing hardware and operation procedures.
- This 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.
- This process allows rapid on-site tailoring of additive-sorbent ratios in order to match the requirements of flue gas changes, such as needed when changing fuels or reducing loads, thus further optimizing the economics.
- This technology reduces the amount of spent sorbents that are disposed.

With the foregoing and other features in view, there is provided, in accordance with the invention, a process for preparing and regenerating carbon sorbents whose activity for mercury capture is promoted by the addition of bromine to the carbon structure .

The invention applies to use of a sorbent in a powdered form that has been injected into a flue gas stream before or after ash particulates have been removed. Examples of the composition of the sorbent are a powdered modified activated carbon prepared by adding Br_2 or HBr plus a second optional component. Alternatively, the sorbent is on a moving contactor consisting of particles or fibers containing the compositions listed in the previous sentence. Either type can be regenerated; the poisoning contaminants from the flue gas are 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 art of adding a reagent (such as peroxide, gold, triiodide, etc.) to the sorbent. The activated carbon sorbent treated with bromine and/or the other 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 incorporated by reference herein.

Although the invention is described herein as a sorbent material, 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.

The construction and method of operation of the invention, however, together with additional objects and advantages thereof will be best understood from the following description of specific exemplary embodiments.

DESCRIPTION OF EXEMPLARY EMBODIMENTS

FIG. 2 is a block diagram illustrating the use of the invention in a coal fueled facility. Of course, the invention can also be used in any other desired type of facility. FIG. 2 shows a boiler for burning pulverized coal. The facility utilizes various devices to clean the exhaust of the boiler. In this example, a baghouse or ESP is used to collect particulates in the exhaust. A scrubber and sorbent bed are also used to remove undesired constituents from the flue gas stream, before being fed to the stack. In the example shown, the sorbent is injected into the flue gas after the boiler. The additive can be injected where desired (e.g., before, after, or within the boiler).

Another advantage of the present invention relates to the use of a feedback system to more efficiently utilize the invention. Where possible, the mercury control technology of the present invention will utilize measurement of mercury emissions as feedback to assist in control of the sorbent injection rate. Tighter control on the sorbent levels in this way will ensure mercury removal requirements are met with the minimal sorbent requirements and therefore the associated costs will be minimized. In one example, the emissions are measured at the stack.

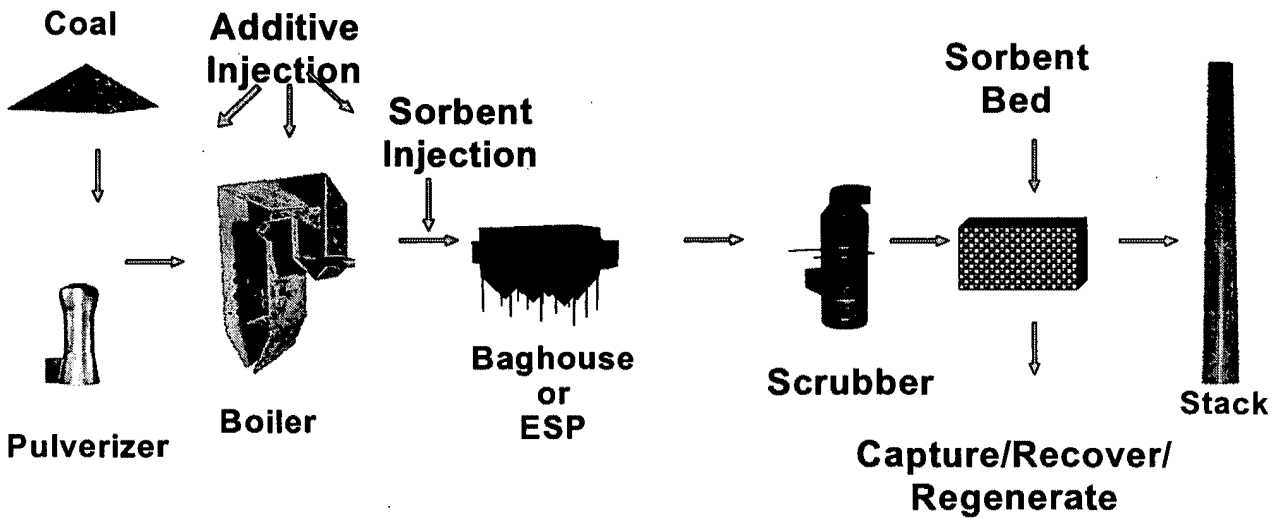

FIG. 2

Carbon Sorbent

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, galium, Sb, Pb, Bi, Cd, Ag, Cu, Zn, or other contaminants, will also react with the oxidation sites generated on the carbon.

According to our model, adding the bromine from the bromine reagent or a proton from a hydrogen halide acid to a basic carbene site on the carbon edge structure forms a carbocation that accept 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.

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A bench-scale procedure based on the above description was used to test the initial activities and capacities of several powdered carbon sorbents, 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. Adding a second component to the solution was demonstrated to improve the capacity of the sorbent.

The carbons were initially tested in a heated bed, where a synthetic flue gas stream containing elemental mercury (11 $\mu g/m^3$) was passed through the bed. Concentrations of total and elemental Hg in the effluent gas were determined using a Sir Galahad mercury CEM. 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 has been treated with 0.1 N HBr, collecting the powder by centrifugation and drying, revealed that the mercury capture activity is 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 indicates that it not only captures mercury at the same level as before (100% capture) but is enhanced such that its capacity is prolonged by several minutes. 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.

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 of ebullated bed of the carbon. The amount of bromine taken up by the carbon ranges (in one example) from <1 to about 30 g per 100 g of activated carbon, depending on the proportions used.

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 carbons were collected in the baghouse at 350E to 375EF. Injection of commercial-grade activated carbon powder (untreated) at a rate of 1.0 lb/Macf resulted in mercury effluent concentrations of 3.8-4.2 $\mu g/m^3$ (representing 62%-58% removal of the mercury from the gas), and at 2.0 lb/Macf, 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-treated carbon.

Addendum: Other References to Hg Sorption

Sorbent Beds with Particulate Removal

Carbon beds have also been utilized for mercury capture in flue gas. The German Patent 34 26 059 describes the use of a very thick carbon bed for treatment of flue gases containing polyhalogenated compounds. Although the system employed at Dusseldorf would also have the potential for Hg removal (for example, see the Streng reference), it is not very practical to use such a thick bed. Spent sorbent is burned and not regenerated. The carbons are not pretreated.

The GE-Mitsui-BF system (Tsuji, K.; Shiraishi, I.; Dague, R.F. Proceedings, Sixth International Symposium, Air & Water Management Assoc., New Orleans, LA, March 10-12, 1993)employs a recirculating carbon bed, where mercury is removed along with acid gases (as ammonium salts) and the carbon is regenerated at high temperatures where ammonium sulfate is decomposed to SO_2 and N_2 and mercury is converted to the elemental form, which desorbs from the sorbent. Attrition of the sorbent results in a significant sorbent cost. The carbons are not pretreated.

Sorbent Injection After Particulate Removal

A process of injecting manganese oxide sorbent particles is described in U.S. Patent Application 2002/0150516. Regeneration is claimed by removal of spent oxide particles from the reaction zone and rinsing with dilute aqueous acid.

Sorbent Beds After Particulate Removal

Of particular interest in designing a mercury control process is to use the sorbent downstream of a particulate control device so the sorbent is not highly diluted with the ash particles. The sorbent could then be more easily regenerated and recycled. The prior art teaches several examples of this type of configuration and sorbent processing.

U.S. Patent 5,607,496 teaches the oxidation of mercury on a metal oxide sorbent bed and subsequent absorption to sorbent. The sorbent bed follows the particulate-removal equipment and, thus, the gas still contains the SO_x and NO_x, which react with the metal oxide sorbent to form the metal sulfates, which poison the bed. High temperatures are proposed to regenerate the bed. However, Hg is only partially removed from the sorbent at temperatures up to 500°C. The sorbents do not work effectively after regeneration using this technique; the problem may be that manganese sulfate formed during the sorption cycle does not completely decompose back to an active manganese oxide form.

U.S. Patent 6,383,981 describes a fixed MnO_2 or Fe_2O_3 bed for removal of Hg from a hydrocarbon stream. No regeneration method appears to be included.

A recent U.S. Patent application (2001/0003116) describes the regeneration of a plate or honeycomb material composed of transition metal oxides that was used for sorption of mercury in flue gas. The claimed process involves heating the sorbent in a reducing gas stream to remove poisons, followed by impregnation with a polyfunctional complex-forming reagent containing

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

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

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

Hg Sorbents for Air or Natural Gas Treatment

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

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

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

Following is a List of Various Background References:

Lancia, A.; Musmarra, D.; Pepe, F.; Volpicelli, G. *Combust. Sci. & Technol.* **1993**, *93*, 277.
Streng, S.; Kassebohm, B. *Fuel Proc. Technol.* **1994**, *39*, 431.
U.S. Patent 6,214,304 Rosenthal
U.S. Patent 4,889,698 Moller
U.S. Patent 4,956,162 Smith
U.S. Patent 5,672,323 Bhat.
U.S. Patent 5,827,352 Altman
U.S. Patent 6,027,551 Hwang
U.S. Patent 5,505,766 Chang
U.S. Patent 5,607,496 Brooks
U.S. Patent 6,383,981 Blankenship
U.S. Patent 4,786,483 Audeh
U.S. Patent 3,194,629 Dreibelbis, et al

U.S. Patent 4,101,631 Ambrsini, et al
U.S. Patent 3,662,523 Revoir
U.S. Patent 4,708,853 Matviya
U.S. Patent 6,258,334 Gadkaree
U.S. Patent 6,136,072 Sjostrom
U.S. Patent Application 2002/0150516
U.S. Patent Application 2001/0003116 Neufert

PATENT APPLICATION SERIAL NO. _____

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PHONE NUMBER:	SENDER'S REFERENCE NUMBER: EER.P0023P
RE: 60/605,640	YOUR REFERENCE NUMBER:

NOTES/COMMENTS:

Re: U.S. Patent Application for:
"SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY"
Applicant: Olson et al.
Serial. No.: 60/605,640
Filed: August 30, 2004
Atty. Docket.: EER.P0023P
Client Ref. No.: 05-001

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Attached are the following:

1. PTO/SB/21 Transmittal (1 page)
2. A corrected Provisional Application Cover Sheet, identifying a government contract (1 page)

Bruce Johnson
37,361

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Express Mail Label No. **EO 904 868 873 US**

INVENTOR(S)

Given Name (first and middle [if any])	Family Name or Surname	Residence (City and either State or Foreign Country)
Edwin S.	Olson	Grand Forks, ND USA
Michael J.	Holmes	Thompson, ND USA

Additional Inventors are being named on the 1 separately numbered sheets attached hereto

TITLE OF THE INVENTION (500 characters max)

SORBENTS FOR THE OXIDATION AND REMOVAL OF MERCURY

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City		State		Zip	
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ENCLOSED APPLICATION PARTS (check all that apply)

☑ Specification Number of Pages 17 ☐ CD(s), Number

☐ Drawing(s) Number of Sheets ☐ Other (specify)

☐ Application Data Sheet. See 37 CFR 1.76

METHOD OF PAYMENT OF FILING FEES FOR THIS PROVISIONAL APPLICATION FOR PATENT

☒ Applicant claims small entity status. See 37 CFR 1.27.

☐ A check or money order is enclosed to cover the filing fees.

☐ The Director is hereby authorized to charge filing fees or credit any overpayment to Deposit Account Number:

☑ Payment by credit card. Form PTO-2038 is attached.

FILING FEE Amount ($): **$80**

The invention was made by an agency of the United States Government or under a contract with an agency of the United States Government.

☐ No.

☑ Yes, the name of the U.S. Government agency and the Government contract number are: U.S. EPA Grant No. R827649-01

[Page 1 of 2]

Respectfully submitted,

SIGNATURE

Date 2-15-05

REGISTRATION NO. 37361 (if appropriate)

TYPED or PRINTED NAME Bruce A. Johnson

Docket Number: EER.P0023P

TELEPHONE 512-301-9900

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TRANSMITTAL FORM

(to be used for all correspondence after initial filing)

Application Number	60/605,640
Filing Date	8/30/2004
First Named Inventor	Olson, Edwin S.
Group Art Unit	
Examiner Name	
Total Number of Pages in This Submission	2
Attorney Docket Number	EER.P0023P

ENCLOSURES ***(check all that apply)***

- ☐ Fee Transmittal Form
  - ☐ Fee Attached
- ☐ Amendment / Reply
  - ☐ After Final
  - ☐ Affidavits/declaration(s)
- ☐ Extension of Time Request
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- ☐ Information Disclosure Statement
- ☐ Certified Copy of Priority Document(s)
- ☐ Response to Missing Parts/ Incomplete Application
  - ☐ Response to Missing Parts under 37 CFR 1.52 or 1.53
- ☐ Assignment Papers *(for an Application)*
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- ☐ Status Letter
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  - corrected Provisional Patent Application Cover Sheet

Remarks

SIGNATURE OF APPLICANT, ATTORNEY, OR AGENT

Firm *or* Individual name	Johnson & Associates Bruce A. Johnson **Customer Number 30163**
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INVENTOR(S)

Given Name (first and middle [if any])	Family Name or Surname	Residence (City and either State or Foreign Country)
Edwin S.	Olson	Grand Forks, ND USA
Michael J.	Holmes	Thompson, ND USA

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FILING FEE Amount ($): $80

The invention was made by an agency of the United States Government or under a contract with an agency of the United States Government.

☐ No.

☑ Yes, the name of the U.S. Government agency and the Government contract number are: U.S. EPA Grant No. R827649-01

Respectfully submitted,

Date 2-15-05

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REGISTRATION NO. 37361 (if appropriate)

TYPED or PRINTED NAME Bruce A. Johnson

Docket Number: EER.P0023P

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