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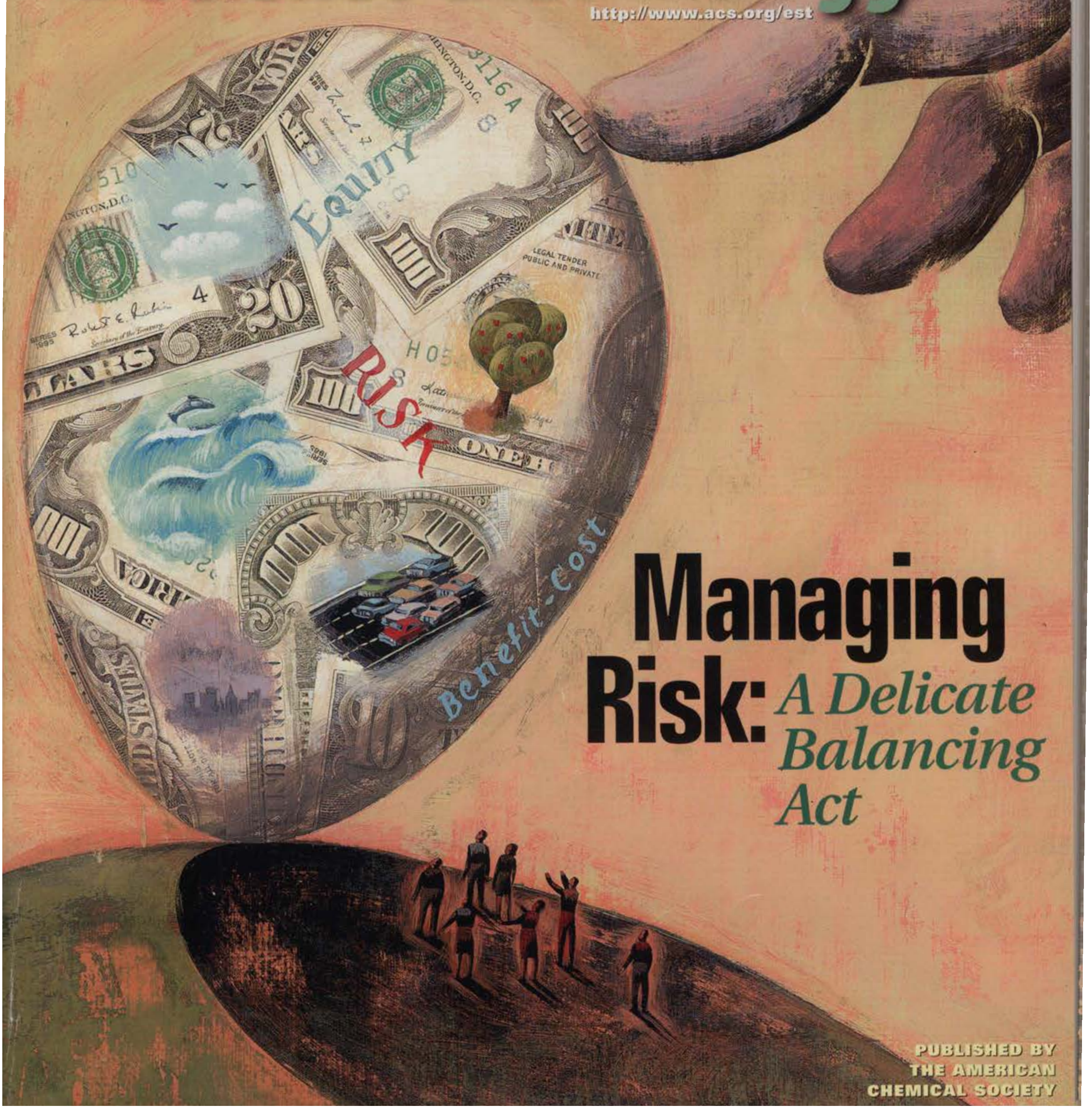

Managing Risk: *A Delicate Balancing Act*

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Research at a Glance

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Cover: Granger Morgan questions current one-size-fits-all risk management practices and suggests that an innovative strategy for accomplishing both benefit–cost and equity objectives should be explored. (Artwork by Curtis Parker)

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Impact of Flue Gas Conditions on Mercury Uptake by Sulfur-Impregnated Activated Carbon

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Novel sulfur-impregnated activated carbons (SIACs) have shown excellent mercury uptake capacity when pure nitrogen was used as a carrier gas. This study investigated the impact of various gas constituents found in a real flue gas on the performance of SIACs. Fixed-bed adsorber tests showed that CO_2 (up to 15%) had no impact on mercury uptake by SIAC, while the presence of O_2 (up to 9%) increased the adsorptive capacity up to 30%. Increase in the amount of oxygen-containing acidic surface functional groups had no impact on mercury uptake, and it is postulated that the enhanced performance was due to the formation of HgO catalyzed by SIAC. Moisture presence (up to 10%) can decrease SIAC's capacity for mercury uptake by as much as 25% due to competitive adsorption and additional internal mass transfer resistance. SO_2 (1600 ppm) and NO (500 ppm) exhibited no impact on mercury uptake by SIAC even in the presence of 10% moisture. Adsorptive capacity of SIAC decreased significantly when the reaction temperature increased from 140 to 250 and 400 °C due to the pronounced exothermic nature of HgS formation, but increasing the empty-bed contact time can partially offset this loss of capacity.

Introduction

Since the Clean Air Act Amendments were enacted in 1990, the U.S. Environmental Protection Agency (EPA) has been focused on evaluating and implementing new regulatory standards to control air toxic metal emissions. Mercury attracted significant attention due to its toxicity (*1*−*3*). The EPA's draft mercury report to Congress in 1995 showed that coal-fired power plants, municipal waste combustors, and medical waste incinerators account for 216 ton/yr of the overall 253 ton/yr of total anthropogenic mercury emissions in the United States (*4*). The EPA's final mercury report released in 1997 (*5*) showed that the total mercury emission from point-sources was lowered to 158 ton/yr. However, coal-fired power plants, municipal waste combustors, and medical waste incinerators still accounted for 75% of the total emission.

Prompted by the possibility of more stringent regulations on mercury, much work has been done or is under way to develop more efficient processes to reduce mercury emissions from point-sources. Traditional pollution control techniques, such as baghouses, ESPs, and wet scrubbers, are able to capture oxidized forms of mercury to some extent (*6*−*8*). However, these devices showed low efficiency for the control of elemental mercury emissions. As a result, a significant amount of elemental mercury leaves the stacks. Researchers (*9*−*11*) have found that solid sorbents, especially chemically impregnated activated carbons, showed greatly improved efficiency for elemental mercury removal that was strongly related to sorbent characteristics (e.g., surface area, chemical composition) and operating conditions (e.g., temperature, residence time, flue gas composition).

Previous studies (*11, 12*) investigated mercury removal efficiency of sulfur-impregnated activated carbons (SIAC) produced at elevated temperatures using pure nitrogen as a carrier gas. These novel adsorbents showed enhanced adsorptive capacity when compared to commercially available products because of a higher content of active sulfur molecules and a larger surface area.

Although using pure nitrogen in these studies was helpful to understand the baseline performance of these sorbents, the real coal-fired power plant flue gas contains other gas constituents such as carbon dioxide (10−15%), oxygen (3−7%), moisture (8−10%), and trace amounts of SO_2 (100−3000 ppm), NO_x (200−600 ppm), and HCl (0−50 ppm) (*8, 13, 14*). It is therefore necessary to investigate the impact of these flue gas constituents on the performance of these novel SIAC adsorbents. A systematic approach was used in this study to evaluate the influence of one gas constituent at a time because the coexistence of several gas components could have compounding impacts and preclude fundamental understanding of their possible effects on mercury uptake. For example, a particular gas constituent may improve the performance of the carbon, while another may deteriorate it, but their simultaneous presence in the carrier gas stream would not allow such observations. Furthermore, certain gases may react with others if they are mixed together, and byproducts may affect the adsorptive capacity of the adsorbent. It is crucial to understand the behavior and impact of each individual gas component. The resulting performance of the sorbent can then be discussed based on the possible molecular interactions among different gas constituents, mercury, sulfur, and activated carbon surface.

Experimental Methods

Column runs for these newly developed SIAC adsorbents were conducted in a fixed-bed reactor charged with 100 mg of 60 × 80 U.S. Mesh size virgin (BPL) or sulfur-impregnated activated carbon (SIAC) and operated at 140 °C using the carrier gas flow rate of 1.0 L/min (unless otherwise noted). Detailed description of sorbent preparation and experimental parameters for column tests is given elsewhere (*11, 12*) and will not be repeated here.

The first step to ensure quality control in this study was to check if any gas constituent interfered with elemental mercury detection by atomic absorption spectrophotometer (AAS). It was determined that CO_2, O_2, and NO had no impact on mercury measurements by AAS. Since water vapor affected the AAS reading, the effluent gas was first passed through a Nafion dryer (Perma Pure, Inc., Toms River, NJ) to eliminate this interference. The gas lines from the moisture generator to the dryer (including the first foot of the dryer tube) were heated to prevent moisture condensation. A hygrometer, Testo 610 (Testo Inc., Flanders, NJ), was used to monitor relative humidity of the carrier gas. The Nafion dryer was able to remove the moisture from the gas stream without

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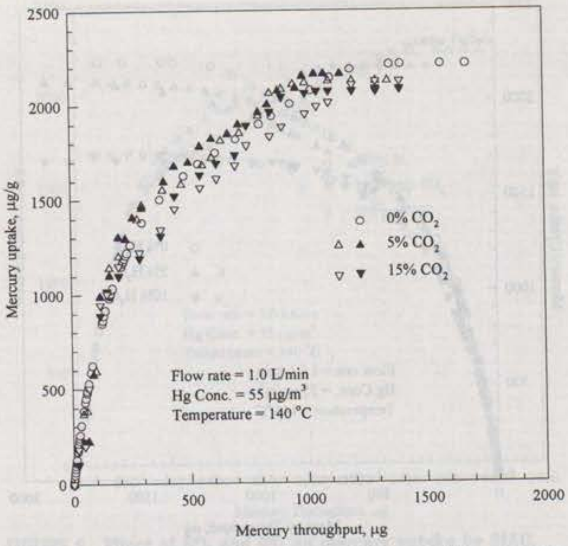

FIGURE 1. Impact of CO_2 on mercury uptake by SIAC.

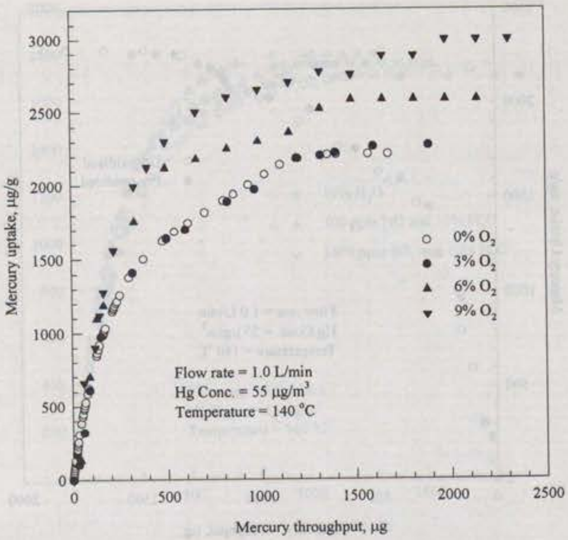

FIGURE 2. Effect of oxygen on mercury uptake by SIAC.

any impact on mercury concentration or speciation in the vapor phase.

It was also observed that SO_2 affected the AAS detection capability, and 0.1 M NaOH solution in a gas-washing bottle was used to scrub SO_2 from the gas stream. It was then also necessary to use the Nafion dryer to remove water vapor entrained in the gas-washing bottle to ensure that AAS adequately detected elemental mercury concentration in the carrier gas.

Additional quality control testing was performed to determine if any of the gas constituents would react with elemental mercury in the gas phase in the absence of carbon surface. Nitrogen flow at 1.0 L/min was first used as a carrier gas, and the empty column was connected to the system. The oven temperature was maintained at 140 °C, and all system parameters were adjusted until the AAS reading indicated an elemental mercury concentration of 55 $\mu g/m^3$. Then, nitrogen was replaced with 1.0 L/min of carrier gas containing one of the major or trace components that will be used in this study. The AAS was monitored continuously for the variation in instrument reading, and impinger trains (*11, 12*) were used to collect the mercury-laden gas for 1 h. The results of the liquid-phase mercury analysis and AAS monitoring confirmed that the elemental mercury concentration in the carrier gas remained at 55 $\mu g/m^3$ for all carrier gas compositions evaluated in this study. For each column run, periodic collection of the effluent by liquid impingers (*11, 12*) was used to verify that no interaction between mercury and different carrier gas constituents occurred even in the presence of the sorbent surface, while combustion of spent sorbents (*12*) was used to verify the mass balance for mercury.

Results and Discussions

Impact of Carbon Dioxide. The results described in the Experimental Section showed that CO_2 did not interfere with elemental mercury measurements and did not react with vapor-phase mercury. Effect of CO_2 on the performance of SIAC was tested at two different concentrations, namely, 5% and 15% (both tests were performed in duplicate), while the remainder was pure N_2 gas.

Figure 1 compares mercury uptake by SIAC in the presence of different CO_2 concentrations. As can be seen from this figure, the adsorptive capacity of SIAC was virtually unchanged regardless of the presence of CO_2. This result indicated that CO_2 behaves like an inert gas during the course of the experiment and does not affect the performance of SIAC.

Impact of Oxygen. The concentration of O_2 in the carrier gas was varied from 0 to 9%, and the resulting mercury uptake is shown in Figure 2. When the concentration of O_2 was increased from 0% to 3%, the mercury uptake capacity of SIAC remained almost unchanged. The overall mercury removal capacity increased by 16 and 33% as the O_2 concentration increased to 6% and 9%, respectively.

Since oxygen is readily chemisorbed by activated carbons to form carbon−oxygen complexes that are important in determining surface reactions and adsorptive behavior (*15*), it was necessary to study the possibility of carbon−oxygen complexes formation during the column tests conducted in the presence of oxygen and their impact on mercury removal.

Both virgin carbon (BPL) and SIAC were tested for the formation of acidic oxygen complexes upon exposure to oxygen using the procedure described by Tessmer et al. (*16*). Preoxidized samples are those prepared by contacting 2 g of the unoxidized samples with a stream of air (flow rate of 1.0 L/min) in a ceramic boat at 140 °C for a period of 7 days. Total acidic surface oxygen content of virgin BPL increased from 445 ± 15 to 578 ± 8 μequiv/g (based on triplicate measurements) as a result of air treatment, while that increase was more pronounced for SIAC (from 130 ± 20 to 620 ± 5 μequiv/g). Lower acidic surface functional group content of unoxidized SIAC as compared to BPL can be explained by outgassing effects during exposure of BPL carbon to a temperature of 600 °C (*16*). The finding that carbon−oxygen complexes formed on SIAC after exposure to oxygen are equal to those formed on preoxidized BPL carbon suggests that a significant portion of the original surface area of BPL carbon remained reactive even after impregnation with sulfur. Sulfur analysis of the SIAC showed that a negligible amount of sulfur (less than 1% of impregnated sulfur) was lost during 7 days of contact with air stream at 140 °C.

Column tests were performed using these unoxidized and preoxidized carbons with pure N_2 as a carrier gas. The breakthrough curves depicted in Figure 3 clearly show that air pretreatment had no impact on the performance of SIAC. Identical observation was made in the case of BPL carbon. In addition, 1% sulfur loss did not reduce the adsorptive capacity of SIAC. These results indicate that air can oxidize carbon surface and increase its acidic surface

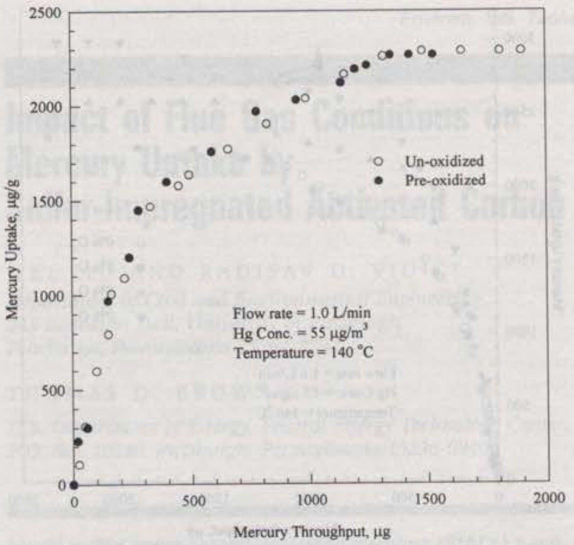

FIGURE 3. Mercury uptake by SIAC before and after oxidation with air.

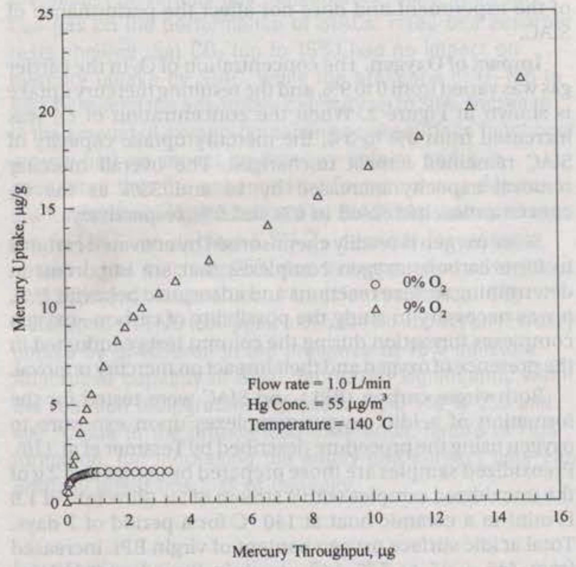

FIGURE 4. Catalysis of mercury and oxygen reaction by BPL.

functional group content, but these changes have no impact on the performance of activated carbon for mercury removal.

It is reasonable to assume that the reaction between O_2 and mercury that is catalyzed by activated carbon surface (*17*) is responsible for the enhanced performance of SIAC in the presence of oxygen (Figure 2), since oxidized mercury is much more adsorbable than elemental mercury. Virgin BPL was tested for mercury uptake using a carrier gas containing 9% oxygen to test this hypothesis. Figure 4 illustrates that virgin carbon showed much higher capacity for mercury uptake (20 µg of Hg/g of carbon) after 4 h of contact in 9% O_2 than in the absence of oxygen (1.5 µg of Hg/g of carbon at saturation). Since virgin BPL can only remove 1.5 µg of Hg/g of carbon in the absence of oxygen, the additional capacity for mercury removal in the presence of oxygen can only be explained by the conversion of mercury to mercuric oxide as there was no reaction between oxygen and mercury in the absence of activated carbon surface.

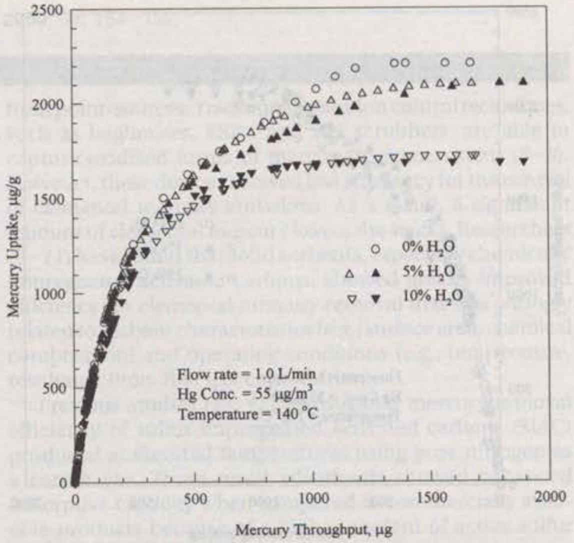

FIGURE 5. Effect of moisture content on mercury uptake by SIAC.

Although, the virgin activated carbon acted as a catalyst for mercury oxidation, it is possible that its catalytic effect could decrease due to the loss of surface area resulting from sulfur impregnation. Surface area of activated carbon covered with sulfur molecules for SIAC would be about 7% of the initial surface area of virgin BPL of 900 m^2/g if it is assumed that sulfur molecules (diameter, $D = 2.08 \times 10^{-10}$ m) (*18*) are impregnated onto the carbon surface in a monolayer. The actual available activated carbon surface area should be higher since the sulfur molecules may form multiple layers on the carbon surface, and it is reasonable to assume that SIAC still has enough active sites to catalyze the reaction between mercury and oxygen as seen for virgin activated carbon.

Impact of Moisture. Figure 5 shows the impact of carrier gas moisture content on mercury uptake by SIAC. The total mercury uptake capacity did not change significantly when 5% moisture was introduced in the carrier gas (based on duplicate test). However, carbon adsorptive capacity decreased as much as 25% when the moisture content increased to 10% (based on duplicate test). Since the adsorptive capacity of carbon did not change at low moisture content, it can be concluded that moisture does not affect the reaction between sulfur and mercury. Therefore, it is postulated that the effect of moisture is related to the adsorption of water by the carbon surface. The carbon surface can bind water molecules to form hydrogen bonds with other molecules (*19*). Higher vapor pressure will increase the amount of adsorbed water. For the 5% moisture in the carrier gas, the capillary condensation may be the dominant process. As the water vapor pressure increased to 10%, water molecules were able to fill the micropores so that isolated water zones merged to block the access to some active sites on the carbon surface and active sulfur molecules, thereby creating additional mass transfer resistance for the adsorption of elemental mercury.

Another factor that could influence the carbon performance is the hydrogen formation due to the dissociation of water induced by carbon. It was reported that hydrogen and CO can be formed if sufficient water vapor pressure is present above the carbon surface in the temperature range used in this study and that hydrogen is preferentially retained by the carbon (*15*). Due to extremely small size, hydrogen molecules can easily reach different size carbon pores to form strong hydrogen–carbon complexes. As a result, the available surface area in the mesoporous region is decreased, and mercury is unable to react with sulfur retained in those pores.

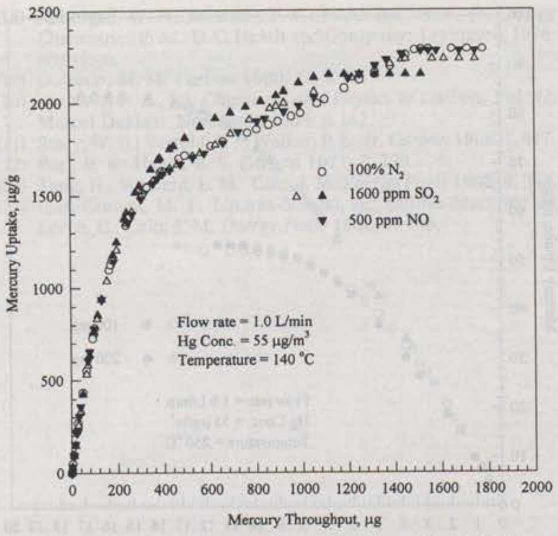

FIGURE 6. Effect of SO_2 and NO on mercury uptake by SIAC.

Impact of Sulfur Dioxide. Figure 6 compares mercury uptake by SIAC in pure N_2 and in 1600 ppm SO_2/N_2 mixture. This test indicated that the mercury uptake capacity of SIAC was not affected by the presence of SO_2. Identical observation was made for virgin BPL (data not shown).

It is well-known that SO_2 could interact with activated carbon via physisorption or chemisorption at relatively high temperatures (>500 °C) and high concentrations (*20–22*) according to the following overall reaction:

$$SO_2 + C \rightarrow CO_2 + 1/2S_2$$

Thermogravimetric analysis (TGA) used to investigate the potential impact of SO_2 on virgin BPL and SIAC revealed that the weight of BPL and SIAC samples remained unchanged even after 5 h of exposure to 3000 ppm SO_2 at 140 °C (data not shown). From the overall reaction, every mole of SO_2 would consume 1 mol of carbon and deposit 0.5 mol of S_2, thereby increasing the net weight of carbon by about 20 g/mol of SO_2 reacted. Based on the experimental conditions used in the TGA test (100 mL/min of 3000 ppm SO_2 in N_2 at 140 °C), the weight increase that could have been observed for the 5-h period was estimated at 3.54 mg. The TGA instrument used in this study would have registered even if only 1/3500 of SO_2 reacted with carbon since the detection limit is 0.001 mg. The results of TGA tests suggest that SO_2 did not react with virgin carbon surface or with impregnated sulfur. This behavior is most likely due to low SO_2 concentration and low reaction temperature that retarded possible reactions.

Impact of Nitric Oxide. Figure 6 indicates that the performance of SIAC did not exhibit a significant change as the carrier gas was switched from pure N_2 to NO/N_2 mixture. Identical observation was made for virgin BPL carbon (data not shown). TGA tests showed that the weight of both BPL and SIAC samples remained unchanged upon exposure to 100 mL/min of 400 ppm NO in N_2 after 5 h of exposure at 140 °C (data not shown).

On the basis of the experimental conditions used in TGA tests, the amount of NO passed through the system in 5 h was about 11.59 mg. Since the detection limit of the TGA instrument used in this study is 0.001 mg, it would have registered if only 1/10000 of NO had been adsorbed by the carbon. Since this was not observed, it is postulated that NO was not be adsorbed by the carbon at the test conditions employed.

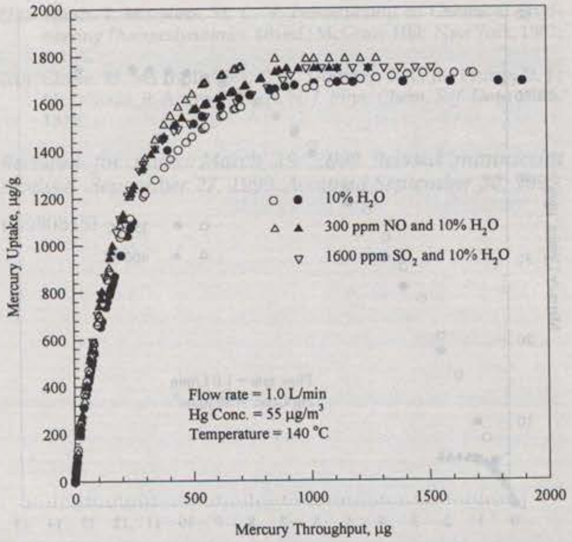

FIGURE 7. Effect of SO_2/H_2O and NO/H_2O on mercury uptake by SIAC.

Several studies found that NO could react with carbon to form various gas products (*23, 24*) based on the following reactions:

$$C + 2NO \rightarrow CO_2 + N_2$$

$$C + NO \rightarrow CO + 1/2N_2$$

$$CO + NO \rightarrow CO_2 + 1/2N_2$$

According to these reactions, carbon weight loss could occur due to gasification. However, this was not observed under the experimental conditions used in this study. One possibility is that the concentration of NO in this study was extremely low (0.0004%) as compared to 1–10% NO used in other studies. Low temperature was another reason for the lack of gasification that was shown to occur at 500–800 °C (*23, 24*). Since the SIAC did not experience any weight loss in the presence of NO, it can be concluded that there was no reaction between NO and elemental sulfur.

Impact of Gas Mixture. Preliminary studies were performed to investigate the combined effects of SO_2 and water vapor and NO and water vapor. Figure 7 shows that the performance of SIAC in the presence of SO_2 (1600 ppm) and 10% moisture or NO (300 ppm) and 10% moisture did not differ very much from the tests conducted in the presence of 10% moisture alone. Even if weak acids (H_2SO_3 or HNO_2) did form on the carbon surface, the low concentration of the acid resulting from the low concentration of SO_2 and NO did not have a major impact on the performance of SIAC, and moisture was the dominant factor influencing the adsorptive capacity of this sorbent.

Impact of Flue Gas Temperature. Mercury uptake by SIAC was evaluated at 250 and 400 °C in order to obtain preliminary information on the performance of this sorbent if it is injected upstream of the air heat exchanger where the flue gas temperatures can be as high as 400 °C. The main advantage of moving the point of sorbent injection further upstream is that the it increases the contact time to as much as 4 s. To accomplish these temperatures in the fixed-bed adsorber, a tube furnace was used as the heating section for the column instead of an oven. Also, stainless steel tubing, which can tolerate high temperatures, was used for the inlet and outlet connections for the reactor in lieu of Teflon tubing. Due to the existence of heat transfer resistance between the

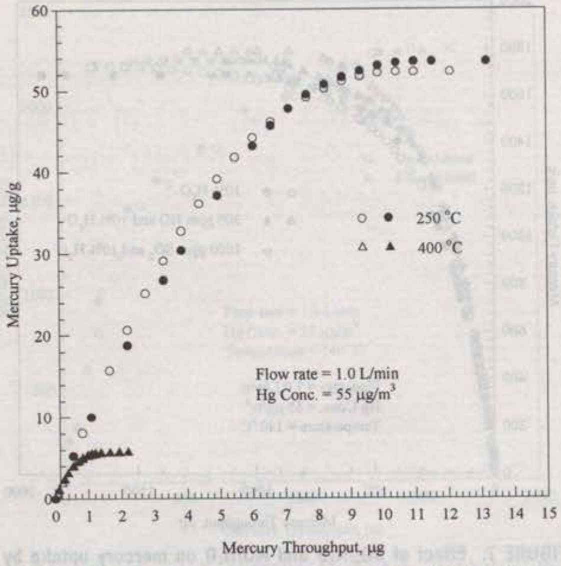

FIGURE 8. Impact of temperature on mercury uptake by SIAC.

TABLE 1. Thermodynamic Parameters of the Reaction between Mercury and Sulfur

temp (°C)	S(l) + Hg(g) → HgS(s)		S(s) + Hg(g) → HgS(s)	
	ΔG (kJ/mol)	K	ΔG (J/mol)	K
25	−82.5	2.88×10^{14}	−82.5	2.83×10^{14}
50	−79.6	7.45×10^{12}	−79.6	7.35×10^{12}
90	−75.1	6.34×10^{10}	−75.1	6.31×10^{10}
140	−69.6	6.30×10^{8}	−69.6	6.32×10^{8}
250	−57.9	6.17×10^{5}	−58.1	6.38×10^{5}
400	−42.7	2.07×10^{3}	−43.6	2.41×10^{3}

heating element and the gas stream, the actual temperature inside the reactor was monitored by a thermocouple. Pure N_2 rather than a mixture of gases was used as the carrier gas so that the results can be compared with previous runs at 140 °C, while the other experimental conditions were the same as before.

Figure 8 shows the rate of mercury uptake by SIAC at 250 and 400 °C. As can be seen from this figure, the performance of SIAC significantly deteriorated at 250 (53.5 μg/g) and 400 °C (5.68 μg/g) as compared to the performance at 140 °C (2600 μg/g). Thermodynamic calculations shown in Table 1 (*25, 26*) indicate that the reaction between mercury and sulfur is highly exothermic and irreversible regardless of the form of sulfur participating in the reaction and that the possibility of HgS decomposition can be ignored. Another important observation is that the ΔG values for the solid and liquid sulfur are almost identical at a given temperature. Therefore, the state change of sulfur does not affect the tendency of the reaction. The values of the equilibrium constant, K, show a drastic decrease with an increase in temperature so that much less HgS can be created at higher temperatures. Since K value at 140 °C is several orders of magnitude higher than K at 250 and 400 °C, significantly larger amount of HgS can be formed at 140 °C, which explains the difference in SIAC performance shown in Figure 8.

Figure 9 shows the impact of empty-bed contact time on SIAC performance at 250 °C. It can be seen that the mercury uptake capacity of this carbon was increased by about 60% when the empty-bed contact time was doubled from 0.011 to 0.022 s. Such behavior suggests that the longer contact time in a fixed bed could at least partially offset the loss of capacity due to the increase in the reaction temperature. However, due to the fact that the initial rates of mercury uptake (slope of the mercury uptake curve at the beginning of the run) are almost identical in both experiments, the impact of longer contact time facilitated by the injection of powdered sorbent upstream of the heat exchanger may be quite limited.

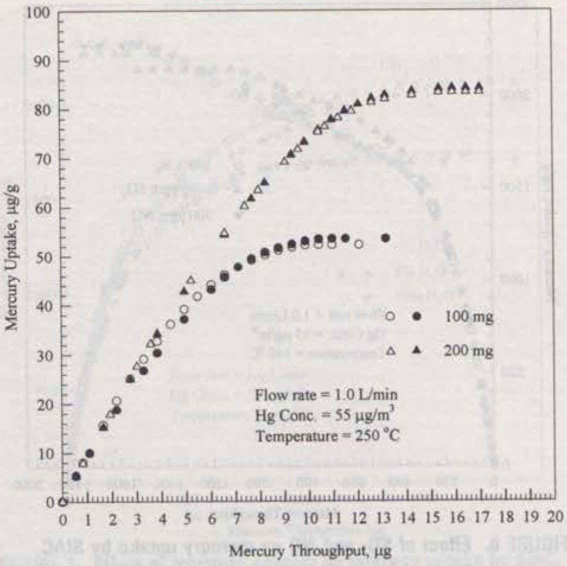

FIGURE 9. Impact of empty-bed contact time on mercury uptake by SIAC at 250 °C.

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