

Modeling Sorbent Injection for Mercury Control in Baghouse Filters: II—Pilot-Scale Studies and Model Evaluation

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ABSTRACT

Activated carbon injection for Hg control in a 500-lb/hr pilot-scale coal-fired furnace equipped with a fabric filter for particulate control was evaluated at different operating conditions. The pilot-scale tests showed that Hg removal was improved at lower temperatures and higher C/Hg ratios. The two-stage mathematical model developed to describe Hg removal using powdered activated carbon injection upstream of a baghouse filter was used to obtain Langmuir isotherm parameters as a function of temperature by fitting the model to a subset of experimental data. The predictive capability of the model was then tested by comparing model calculations with additional experimental data from this system obtained using different operating temperatures and sorbent to Hg ratios. Model predictions were in good agreement with experimentally measured Hg removal efficiency. Based on the

model predictions, Hg removal in the duct appears to be limited and higher C/Hg ratio, lower operating temperature, and longer cleaning cycle of the baghouse filter should be utilized to achieve higher Hg removal in this system.

INTRODUCTION

Environmental control agencies and researchers have become increasingly concerned with the mobilization of trace elements to the environment from fossil-fuel burning and solid-waste incineration. Mercury is the trace element of particular concern because, during combustion, most of the Hg present in the feed stream is transferred into the vapor phase because of its high volatility. There is considerable evidence in the literature that currently used pollution abatement technologies (flue gas desulfurization, control of NO_x and SO_x emissions, and particulate control devices) are not capable of controlling elemental Hg emissions to a significant degree.¹

Once discharged to the atmosphere, Hg persists in the environment and creates a long-term contamination problem. Furthermore, well-documented toxicity and bioaccumulation of Hg require strict control of Hg emissions from coal-fired power plants. In December 2000, the U.S. Environmental Protection Agency (EPA) announced its determination to regulate Hg emissions under the Clean Air Act Amendments of 1990 (Title III, Section 112). EPA is slated to propose regulations by December 2003 and promulgate them by December 2004, with full compliance expected by the end of 2007.

Direct injection of activated carbon into the flue gas stream has been proposed as a relatively simple approach for controlling Hg emissions.² Injected activated carbon binds the vapor-phase Hg through physical adsorption or

IMPLICATIONS

The mathematical model developed in this study offers significant benefits in estimating appropriate design and operating strategies that would lead to efficient and economic control of Hg emissions from coal-fired power plants and utility boilers. Most of the kinetic parameters were estimated from literature correlations and manufacturers' specifications, while adsorption equilibrium parameters were determined by fitting the model to a set of experimental data obtained from a pilot-scale coal combustor system. The predictive capability of the model was demonstrated using experimental measurements on the same pilot-scale system. Model calculations suggest that Hg removal in the duct is minimal and that higher C/Hg ratio, lower operating temperature, and longer cleaning cycle of the baghouse filter should be utilized to achieve higher Hg removal.

chemisorption and is collected in downstream particulate collection devices, such as fabric filters or electrostatic precipitators. Several bench,^{3–5} pilot,^{6–10} and full-scale studies^{11–13} examined the influence of carbon type, carbon structure, carbon surface chemistry, injection method (wet or dry), amount of carbon injected, and flue gas temperature on Hg removal. Inconsistencies in the data reported in these studies suggest that a wide variety of factors may influence the Hg removal. These factors potentially include Hg species being removed (oxidized versus elemental), flue gas composition, process conditions (e.g., temperature), sorbent characteristics, and presence of other active surfaces (e.g., fly ash).

Because activated carbon can be collected effectively in existing particulate control devices, direct activated carbon injection has several potential advantages over spray dryer adsorption and wet scrubbing processes, including (1) simpler operation; (2) lower operational cost; (3) lower capital cost; (4) no wastewater problem; (5) simpler waste disposal; (6) easier maintenance and repair; and (7) greater efficiency for Hg removal. Based on the success of field-scale trials conducted on a number of municipal solid waste (MSW) incinerators, it was anticipated that powdered activated carbon injection into the flue gas with subsequent collection at the fabric filters can also be used for the control of Hg emissions from coal-fired power plants. However, because Hg concentrations in MSW incinerator flue gas (200–1000 µg/m³) are 1–2 orders of magnitude higher than for coal-fired power plants (2–20 µg/m³), as well as other differences in process conditions (e.g., HCl content, flow rate, and SO₂ and NO_x concentration), C/Hg ratios required for effective control of Hg in MSW flue gas are an order of magnitude lower than those necessary to achieve similar Hg removals in coal combustors.¹⁴ Preliminary cost estimates for the control of Hg emissions from coal-fired power plants using powdered activated carbon injections range from \$14,400 to \$38,200 per lb Hg removed.² The use of activated carbon injection for Hg control, therefore, requires further optimization to make this promising technology viable in practice and to reduce severe environmental problems associated with Hg emissions. The objective of this study was to demonstrate that the model described in Part I of this article can be used to predict Hg removal by powdered sorbent injection in combination with a baghouse filter and develop optimal operating strategies for this system to maximize Hg removal efficiency.

PILOT-SCALE STUDIES

The 500-lb/hr furnace system, shown in Figure 1, consists of a wall-fired pulverized coal furnace equipped with a water-cooled convection section, a recuperative air heater, a spray dryer, a baghouse, and associated ancillary

equipment (fin-fan coolers, surge tanks, coal hoppers, blowers, pumps, etc.).^{15,16} The 500-lb/hr combustor is an indirect-fired unit. Coal is first pulverized offline in the Williams roller mill and is then transported through a series of hoppers before it is fed by an Acrison weight-loss differential feeder to the combustor. The combustor is a wall-fired, dry bottom type that is capable of firing both coal and natural gas. The overall system operating performance is characterized by online temperature readings, flow measurements, and four separate banks of continuous gas analyzers (O₂, NO_x, CO, SO₂, and CO₂).

The flue gas from the combustor flows to a convective section, the secondary air preheater, through a spray dryer, a sorbent injection duct (SID) test section, and a baghouse. The spray dryer was only operated during tests involving water injection for direct cooling (i.e., humidification tests). The SID and baghouse were heat-traced to minimize heat losses downstream of the spray dryer and a wide range of flue gas temperatures at the spray dryer inlet, SID, and baghouse can be obtained.

All pilot-scale tests were performed using Norit Darco FGD (Norit Americas, Inc.) activated carbon that was injected at two different locations along the SID. This allows for a range of sorbent in-duct residence times relative to the baghouse and SID gas sampling locations. The sorbent injection system consists of a hopper, a screw feeder, a scale, an eductor, and a compressed air line. The fly ash and the injected sorbent are collected in a 6-ft i.d. cylindrical pulse-jet baghouse that contains 57 bags arranged in nine rows. The Goretex Nomex bags are 8 ft long and 4.5 in. in diameter. The baghouse bags are cleaned (pulsed) with 80-psi air either when the preset pressure drop is exceeded or at regular time intervals. The dislodged fly ash and sorbent are collected in a lined, 55-gal drum, which is weighed and sampled.

The major operating variables investigated include sorbent injection rate (sorbent/Hg mass ratio) and baghouse temperature. The sorbent injection rate can be varied without affecting other pilot unit conditions while the baghouse temperature is adjusted by changing the operation conditions of the pilot unit. Heat duty of the secondary air preheater or the coal-firing rate were varied to achieve three different baghouse temperatures (250, 270, and 300 °F). Recent tests used humidification to control baghouse temperature and to examine the effects of water concentration in the flue gas on sorbent performance. Water for the humidification was added through nozzles installed in the spray dryer unit. The water injection rate was set to achieve a temperature of either 250 or 200 °F. In addition to temperature, baghouse parameters that can affect Hg removals include the fabric material, air-to-cloth ratio, and pulsing frequency. The average baghouse operating conditions were

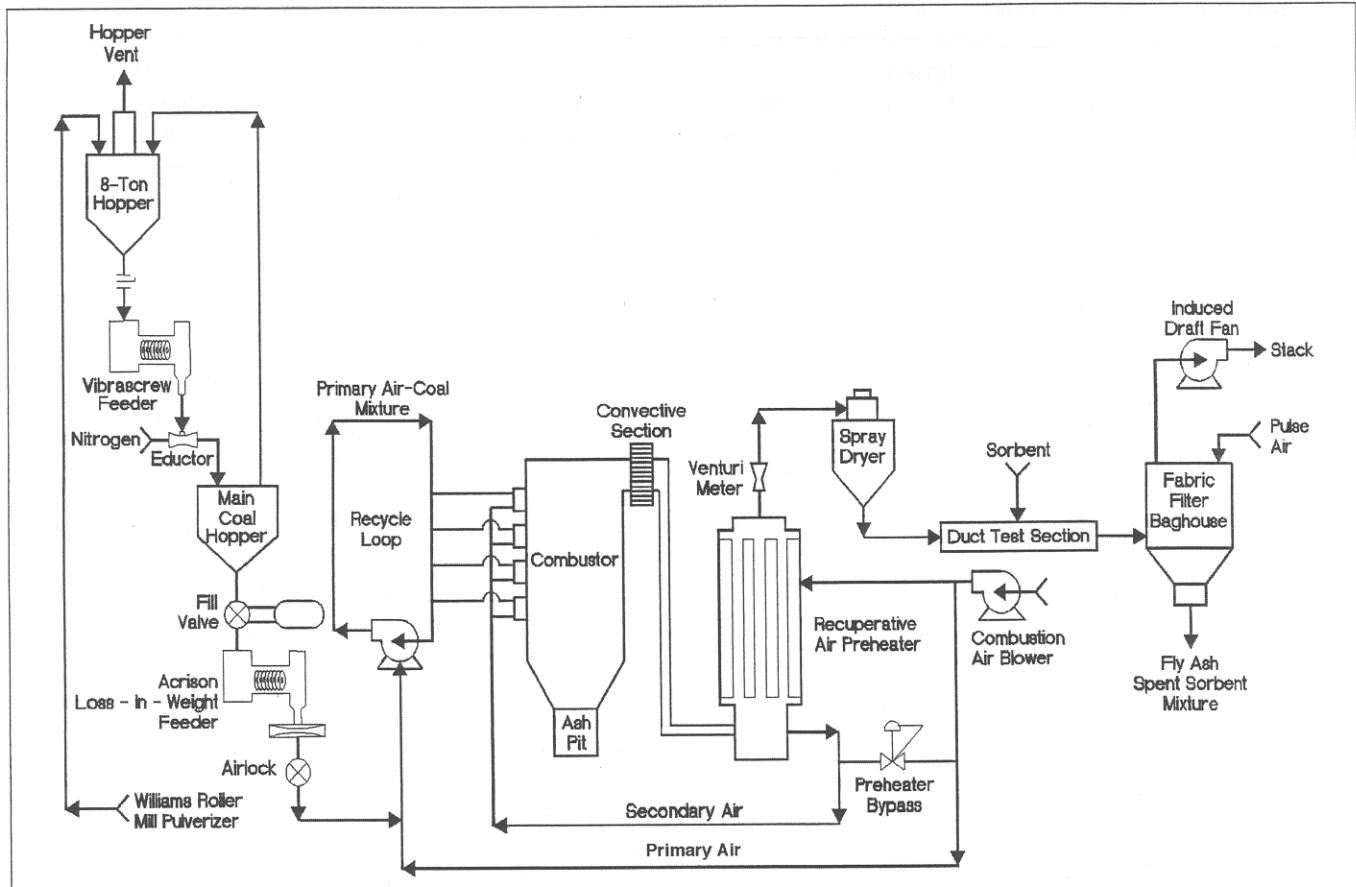


Figure 1. The NETL 500-lb/hr coal combustion test facility.

not varied for the tests described here, but air-to-cloth ratio varied with changes in coal-firing rate.

All Hg removals and material balances were calculated from flue gas measurements using either EPA Method 101A¹⁷ for total Hg or the Draft ASTM Method,¹⁸ also known as the Ontario-Hydro (O-H) method, for total and speciated Hg. These wet-chemical methods have been found to give good, repeatable results on this unit.^{15,16}

RESULTS AND DISCUSSION

Table 1 shows the results of pilot-scale tests, including the average temperature in the system, influent Hg concentration, flue gas flow rate, activated carbon injection rate, and average Hg removal in the system. It is important to note that Hg concentrations and removal efficiencies reported in this table represent steady-state results based on the 1- or 2-hr average sampling period that is required for the O-H method. Mercury mass balances around the baghouse ranged from 77 to 136%, while the mass balance around the entire system averaged 83%.¹⁵

It is clear from the data in Table 1 that the target temperatures of 250, 270, and 300 °F were accomplished reasonably well in these pilot-scale tests. C/Hg ratios of up to 16,300 (g/g) were tested in this study, and Hg removal ranged from negligible to above 90%. As expected, Hg

removal at a particular baghouse temperature increases with increasing sorbent-to-Hg ratio. Also, Hg removal efficiency increases with decreasing baghouse temperature for a constant injection ratio. Data sets 1 and 23, and 5 and 17, showed approximately 10 and 30% Hg removal, respectively, without activated carbon injection. Such behavior can partially be explained by the presence of unburned carbon in fly ash.¹⁶ Data sets 9 and 10 are the only outliers among the 23 data sets shown in Table 1 because a negative Hg removal was measured for relatively low C/Hg ratios of approximately 2000.

Model Calibration

To use the model to describe the system, some parameters were extracted directly from the experiments. In the absence of values for other parameters, values from the literature or those obtained directly from the manufacturer were used. Table 2 shows a list of the parameters used in the model and their sources. Using these parameters, the Langmuir isotherm constants (q_{\max} and b) at a particular average temperature were obtained by fitting the mathematical model simultaneously to the data sets in the pilot-scale studies. The sum of the squares of the differences between the average removal predicted by the

Table 1. Mercury removal from the pilot-scale tests.

Data Set	Temp. (°F)	Influent Mercury ($\mu\text{g}/\text{m}^3$)	Flow Rate (m^3/sec)	AC Injection Rate (g/sec)	C/Hg Ratio (g C/g Hg)	Time in Flight (sec)	Percent Mercury Removal
1	294	2.86	1.47	0.0000	0	N/A	12.2
2	294	2.76	1.53	0.0485	11476	2	84.1
3	265	3.01	1.50	0.0538	11909	2	88.7
4	268	3.05	1.41	0.0288	6701	2	68.2
5	296	2.94	1.46	0.0000	0	N/A	31.0
6	296	3.11	1.49	0.0131	2840	2	42.8
7	296	2.94	1.58	0.0276	5916	2	65.6
8	270	3.30	1.48	0.0144	2964	2	62.7
9	275	2.96	1.20	0.0071	1984	2	(3.7)
10	306	2.96	1.27	0.0081	2157	2	(5.4)
11	244	3.31	1.08	0.0267	7454	2	90.0
12	250	2.32	1.05	0.0147	6065	2	87.5
13	250	2.06	1.10	0.0149	6553	2	79.6
14	200	2.26	1.08	0.0149	6099	2	96.5
15	262	5.31	1.00	0.0146	2756	0.5	80.8
16	301	4.25	1.05	0.0271	6075	0.5	73.2
17	264	5.36	0.92	0.0000	0	N/A	32.8
18	271	4.24	0.97	0.0542	13177	0.5	96.7
19	271	4.14	1.00	0.0551	13310	2	92.5
20	272	3.77	1.01	0.0621	16248	0.5	91.8
21	272	4.24	0.99	0.0294	6997	0.5	87.7
22	271	4.43	0.93	0.0290	7014	2	86.2
23	270	4.41	0.95	0.0000	0	N/A	10.2

model and the data sets was minimized by varying q_{\max} and b within a simulated annealing algorithm.¹⁹

Three data sets within a reasonably close temperature range were used to obtain q_{\max} and b from the experiments.

Table 2. Parameters used for the model.

Parameter	Value
q_{\max}	Estimated from model fit
b	Estimated from model fit
T	Pilot test data
c_0	Pilot test data
Q	Pilot test data
\dot{m}_c	Pilot test data
r_p	Pilot test data
d_p	150 Å (from Hsi et al. ²⁵)
τ_p	7.5 (from Yang ²⁶)
ϵ_b	0.7 (from Bush et al. ²⁷)
ϵ_{cb}	Pilot test data
Time in duct	Pilot test data
Cleaning cycle interval	Pilot test data
Fraction bed cleaned/cycle	1/9 (assumed)
A	49.9 m^2 (from manufacturer)
ρ_p	2.04 g/cm^3 (from manufacturer)
ϵ_p	0.67 (from manufacturer)

Table 3 shows the data sets used and the corresponding values for the isotherm constants, and Figure 2 shows the model fits to the data groups based on the parameters in Table 3. The jagged nature of the model fit is caused by the intermittent cleaning of the baghouse, with the intermittent drop in the fractional removal indicating when the baghouse was cleaned. Different data sets had different cleaning intervals. In all the data sets, it was assumed that 1/9 of the total filter area in the baghouse was cleaned in each cleaning cycle. The average removal from the pilot test is also plotted in the figure. Mercury analyses during the pilot tests were not sufficiently frequent to clearly show the jagged nature predicted by the model.

The model shows that the removal of Hg in the ductwork is minimal. The immediate increase in the removal efficiency after the ductwork is a result of the contribution

Table 3. Isotherm constants obtained at different temperatures.

Data Group	Temperature (°F)	Average		
		Data Sets	q_{\max} ($\mu\text{g}/\text{g}$)	b ($\text{m}^3/\mu\text{g}$)
1	248	11, 12, 13	70714	0.107
2	271	8, 19, 22	2909	7.02
3	295	2, 6, 7	496	20

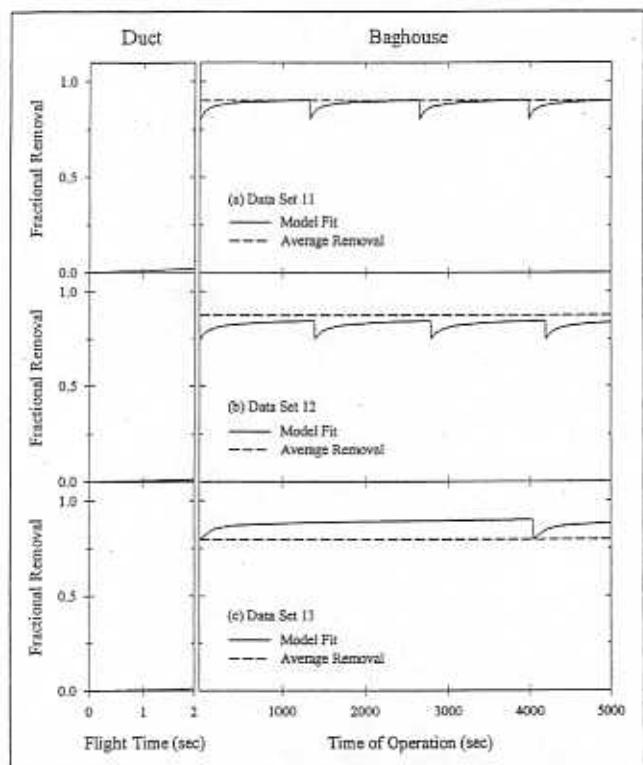


Figure 2. Average Hg removal and model fit for (a) data set 11, (b) data set 12, and (c) data set 13.

of the various fractions of the baghouse filter in Hg removal. The fraction of the filter that was just cleaned would have a low removal efficiency because the filter

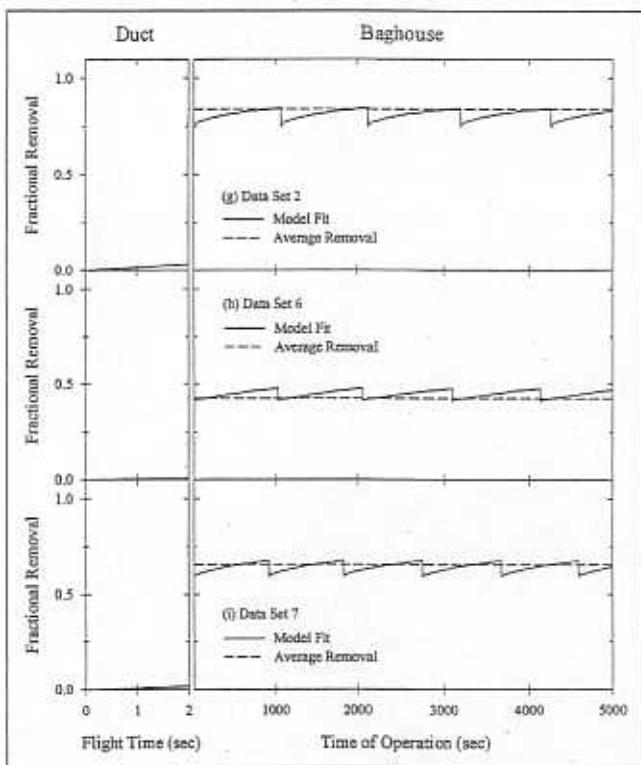


Figure 2. (cont.) Average Hg removal and model fit for (g) data set 2, (h) data set 6, and (i) data set 7.

cake had not yet been formed, while the fraction of the filter with a relatively mature filter cake would already be effectively removing Hg from the flue gas. The resulting composite average contribution of all the fractions of the bed to the Hg removal efficiency is plotted in Figure 2.

Difficulties were encountered when obtaining the isotherm constants from the first and third data sets shown in Table 3. In the first data group, the optimum values of the isotherm constants lay in a very narrow trench where values of the constants varied within 2-3 orders of magnitude and were within 1% of the minimum error. The minimum error was obtained with a low value of b ($8.5 \times 10^{-4} \text{ m}^3/\mu\text{g}$) and a high value for q_{\max} ($8.4 \times 10^6 \mu\text{g/g}$). Because the maximum possible bulk concentration would be the influent concentration (from Table 1, the maximum c_0 is $5.36 \mu\text{g/m}^3$), the search for the optimum tended toward a linear isotherm. This is reasonable because the adsorptive capacity of activated carbon for Hg is higher at low temperatures, and the actual capacity can be parametrically described as a linear isotherm. In contrast, a high value of b ($>100 \text{ m}^3/\mu\text{g}$) was obtained for the optimum in the third data group, indicating that the search for the optimum parameters tended toward a constant isotherm. In this case, a constant isotherm would indicate immediate saturation and is unreasonable. To avoid having a different isotherm to describe the adsorption capacities at different temperature ranges and to obtain a continuous and reasonable variation of

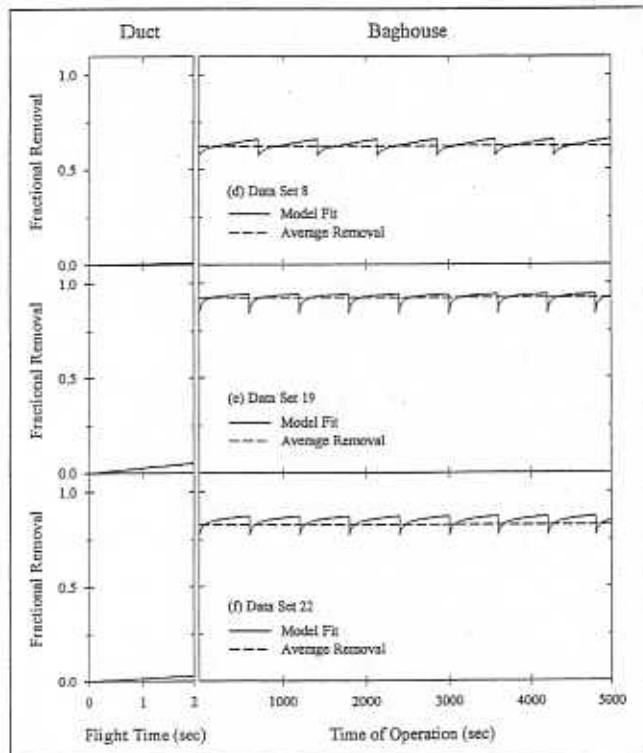


Figure 2. (cont.) Average Hg removal and model fit for (d) data set 8, (e) data set 9, and (f) data set 22.

the isotherm parameters as a function of temperature, the lowest q_{max} and the highest b were selected within the error trench that did not vary by more than 1% of the minimum error for the first data group, and a maximum value of $b = 20 \text{ m}^3/\mu\text{g}$ was arbitrarily assigned for the third data group. The final isotherm constants corresponding to these cases are listed in the table.

A comparison of the model fits to the actual average removal is shown in Figure 3. The figure shows that the model is in good agreement with the average removal for the selected data sets. This demonstrates that the model results were reasonable despite the assumptions made regarding the isotherm parameters during model calibration.

The isotherm parameters and an Arrhenius relationship describing the variation of these parameters with temperature are shown in Figure 4. An increase in temperature results in a lower capacity of the carbon for Hg, which is characteristic of an exothermic adsorption process and is consistent with earlier findings.^{3–6,20} The Langmuir coefficient, b , increased with temperature. This coefficient can be conceptualized as the ratio of the kinetic coefficient for adsorption to the kinetic coefficient for desorption. It is possible that with an increase in temperature, the kinetic coefficient for adsorption increased but the kinetic coefficient for desorption did not increase with the same magnitude. Such variations in the kinetic coefficients with temperature was reported by Karatza et al.^{21,22} for another Norit carbon. Such behavior can also be explained by the irreversible adsorption of Hg on activated carbon^{23,24} because the rate constant of the forward reaction should increase with temperature, as is the case for any chemical reaction, while the rate constant for the reverse reaction (desorption) remains infinitely small. Subsequently, the equilibrium constant b , which represents the ratio of the two rate constants, should increase with temperature. At a temperature of 135 °C (275 °F) and

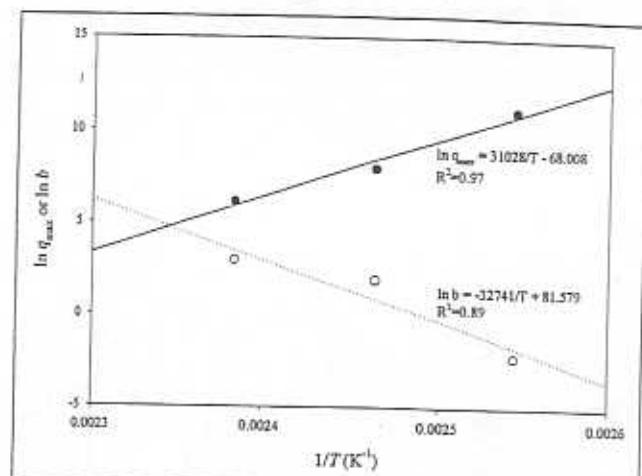


Figure 4. Variation of Langmuir isotherm parameters with temperature.

a Hg concentration of 59 μg/m³, Hsi et al.²⁵ reported the capacity of Norit Darco FGD of 2566 μg/g, which is reasonably close (within 17%) to the capacity calculated for that temperature using the data in Figure 4 (3007 μg/g).

Model Predictions

Figure 5 shows the model predictions for all the data sets (including the data sets used for model fitting) accounting for the variation of the isotherm coefficients with temperature. The model reasonably predicts the baghouse performance with the exception of a few data sets. Because data sets 1, 5, 17, and 23 did not have any carbon injection, the model predicted zero removal of Hg from the baghouse. However, 10–33% removal was observed experimentally, indicating that the fly ash itself²⁴ or unburned carbon¹⁶ may have contributed to Hg removal. In another case, because data sets 9 and 10 had the lowest carbon injection rates among all data sets, the model predicted modest removal efficiencies. However, the actual experimental removal efficiencies were negative. It

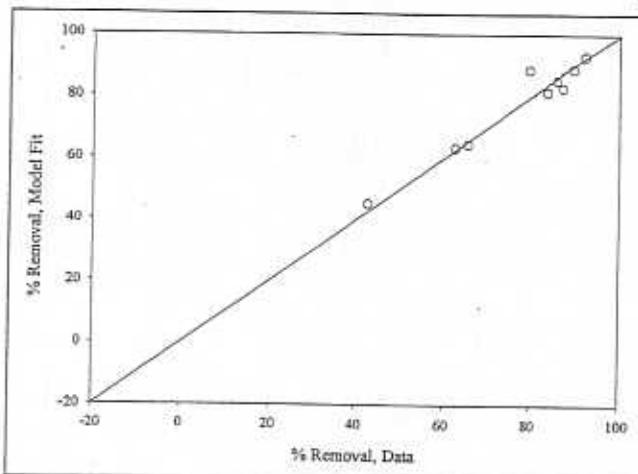


Figure 3. Comparison of the average Hg removal as calculated with the model fits to the average Hg removal data.

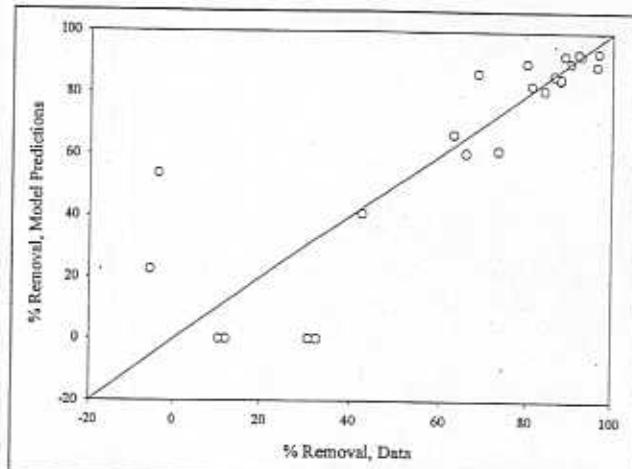


Figure 5. Comparison of the average Hg removal as predicted by the model to the average Hg removal data.

would be expected that the removal efficiencies should at least be equal to the removal efficiencies associated with the fly ash (i.e., 10–33%). Reasons for this discrepancy are not clear at this time.

From Table 3 and Figure 4, it is clear that the temperature will significantly impact the removal of Hg from the baghouse. Another important process parameter that impacts the removal of Hg and is more easily controlled is the carbon injection rate. Figure 6 shows the C/Hg ratio for the experiments and the model. As expected, higher carbon doses result in greater removal of Hg in this system. The model predictions are also in agreement with the general trend of experimental data.

To further illustrate the effects of temperature and the C/Hg ratio, the effects of varying these parameters on the calculated average Hg removal is shown in Figure 7. The conditions for this figure are the same as the base conditions in the first paper of this series (see Table 1) except that the isotherm parameters were varied with temperature using the equations in Figure 4. The range of temperature and C/Hg ratios are typical of the experimental conditions in the pilot-scale tests and those that could be expected in full-scale operations. A decrease in the removal efficiency is observed at high temperatures because the adsorptive capacity at higher temperatures is lower. Increasing the carbon addition (hence, increasing C/Hg ratio) will increase the removal efficiency at any temperature. The magnitude of the improvement in removal efficiency with C/Hg ratio is more pronounced at the higher temperatures where the carbon is more capacity-limited. At lower temperatures, the system is more transfer-limited and the removal efficiency is less sensitive to the mass of carbon added.

The impact of the cleaning cycle and the fraction of the bed cleaned per cycle is shown in Figure 8. A longer cleaning cycle results in a longer detention time of the particles on the fabric filter, which allows the carbon

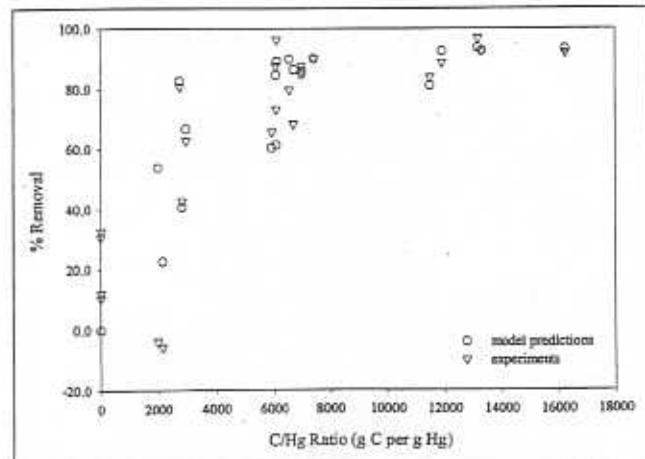


Figure 6. Effects of the C/Hg ratio on the average Hg removal for the NETL data sets.

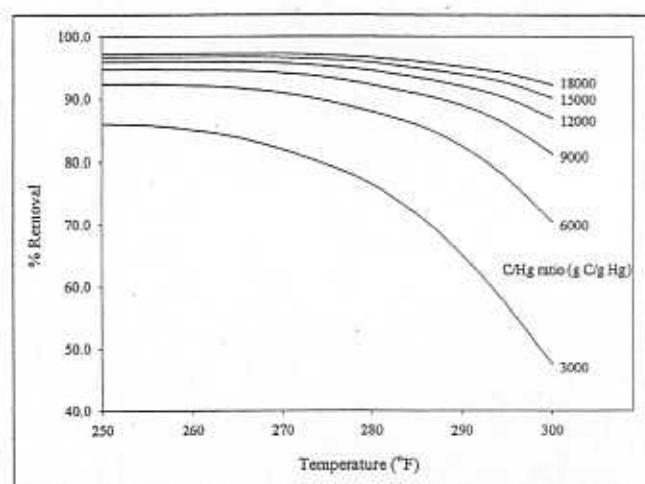


Figure 7. The effects of baghouse temperature and carbon dose (expressed as the C/Hg ratio) on Hg removal efficiency from the baghouse filter.

particles to adsorb more Hg from the bulk solution. This is different from a traditional fixed-bed adsorber because fresh, lightly loaded carbon particles are continuously being added to the influent of the growing bed where the Hg concentration is the highest. The Hg in the bulk phase decreases as it passes through the bed and never achieves the breakthrough observed in a traditional fixed-bed adsorber. As the fabric filter is divided into different fractions for cleaning to maintain the pressure drop across the baghouse, lower fractional cleanings further allow the particles in the "uncleaned" section of the bed to uptake more Hg. The ideal case would be to have long cleaning cycles and low fractional bed cleaning, with the limits dictated by the allowable pressure drop across the baghouse filter. Further studies have to be performed to evaluate the impact of the pressure drop on the Hg removal performance from the baghouse filter.

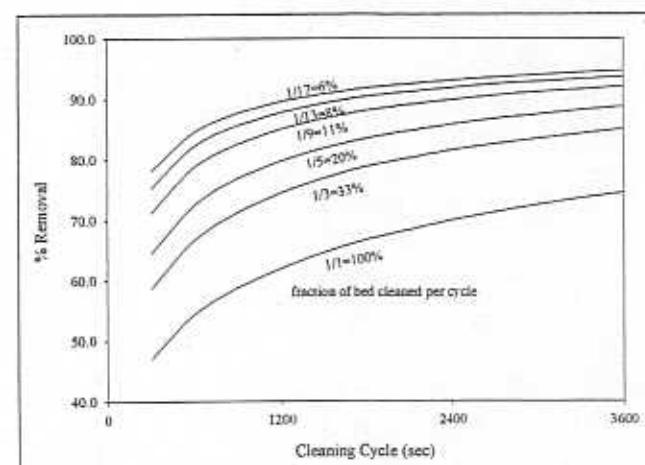


Figure 8. The effects of the cleaning cycle and the fraction of the baghouse cleaned per cycle on the calculated average Hg removal efficiency from the baghouse filter.

CONCLUSIONS

The two-stage mathematical model was used to obtain Langmuir isotherm parameters for Hg on Norit Darco FGD sorbent at different temperatures. An Arrhenius relationship adequately quantified the variation of these parameters with temperature. The model reasonably described Hg removals measured on the pilot-scale system operated by the National Energy Technology Laboratory (NETL) and was used to evaluate expected trends associated with a change in the process conditions (i.e., gas flow rates, Hg concentrations, temperature, carbon loading, etc.). Based on the model predictions, Hg removal in the duct appears to be limited and lower operating temperature, higher C/Hg ratio, and longer cleaning cycle of the baghouse filter should be utilized to achieve higher Hg removal in this system.

The role of the fly ash in Hg removal is not well understood. The experimental data show conflicting results where Hg removal efficiencies between 10 and 33% were observed without carbon injection but negative removal efficiencies were observed with low C/Hg ratios. Thus, it is critical that the adsorption capacities (and the relevant transport parameters) of the fly ash and unburned carbon be characterized. The model developed in this work can be used to estimate the isotherm parameters and predict the performance on only one adsorbent. However, if the fly ash or unburned carbon exhibit significant adsorption capacities, the model needs to be revised to account for the presence of more than one adsorbent.

The current model accounts for the presence of only one Hg species in the vapor phase. However, it is likely that both elemental and oxidized Hg species would exist in full-scale systems. Therefore, the existing model may have to be revised to account for the presence of more than one Hg species in the system and include competitive adsorption features. Unfortunately, analytical techniques and continuous emission monitors that could reliably distinguish and accurately measure two possible Hg species are still under development.

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