

# RATE STUDIES ON VAPOR-PHASE MERCURY UPTAKE BY VARIOUS ACTIVATED CARBONS

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## Introduction

Direct injection of powdered activated carbon into the flue gas stream represents a relatively simple approach for controlling mercury (Hg) emissions from coal-fired power plants. Injected activated carbon binds the vapor-phase mercury through physical adsorption and/or chemisorption and is collected in downstream particulate collection devices, such as FFs or ESPs. According to several bench-scale studies (1-4), sulfur (S) impregnated activated carbons exhibited significantly greater Hg removal capacities than virgin activated carbons. The main focus of this study was to evaluate the influence of temperature, Hg concentration and activated carbon type on the rate of mercury removal (uptake kinetics).

## Experimental

Adsorption experiments with vapor-phase elemental mercury were conducted using BPL, HGR, and BPL-S activated carbons. BPL (virgin bituminous coal-based carbon) and HGR (commercially available sulfur impregnated activated carbon) were supplied by Calgon Carbon Corporation (Pittsburgh, PA). BPL-S carbon was prepared by impregnating virgin BPL carbon with elemental sulfur in a pure nitrogen atmosphere at 600 °C. The sulfur contents of HGR and BPL-S were 9.7% and 10.0%, respectively. Detailed description of the analytical procedures used in this study is available elsewhere (5) and will not be repeated here.

Schematic representation of the experimental setup used in this study is shown in Figure 1. Differential-bed reactor (3/4 inch I.D. stainless steel column) was charged with 100 mg of 170x230 U.S. Mesh size activated carbon and operated in a downflow mode. In order to prevent damage of the system components due to high temperature in the column effluent, the recirculation loop was first passed through a condenser (33-inch long plastic hose supplied with a stream of cold water) to cool the effluent gas. A 10 L polyethylene carboy served as a reservoir to provide larger system volume (the total volume was 13 L).

Each adsorption experiment was performed in two phases: open system and closed system. During the open system phase of the experiment, the path between Valve ①

and Valve ② was closed and 1 L/min of mercury-laden nitrogen gas was directed through the system during a two-hour period to equilibrate mercury vapor throughout the entire system. After the open system phase, the path between Valve ① and ② was opened and the system was converted into a closed system. Prior to initiating the column run, the compressor was turned on and the mercury vapor in the closed system was recirculated for 6 minutes while bypassing the adsorption column. Mercury vapor was then directed through the carbon column and elemental mercury concentration in the system was continuously monitored by AAS.

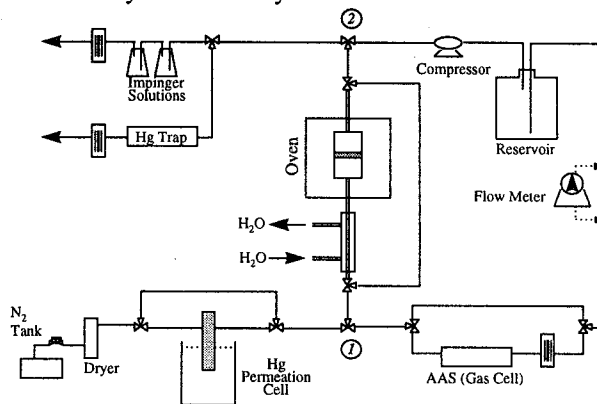


Figure 1. Experimental System

## Results and Discussion

### Mercury Adsorption by BPL Carbon

Results of mercury adsorption experiments with BPL carbon are shown in Figure 2. For the mercury concentration of 110  $\mu\text{g}/\text{m}^3$ , an increase in gas temperature showed little impact on the rate of mercury uptake, which is probably due to the fact that equilibrium was attained almost instantaneously for this low mercury concentration. On the other hand, for the higher initial vapor-phase mercury concentrations, higher temperatures resulted in lower overall mercury uptake, which is characteristic of the exothermic adsorption process. Higher temperatures also resulted in faster kinetics of mercury adsorption by BPL carbon since stable mercury levels were achieved after a shorter period of time when compared to the test conducted at 25 °C.

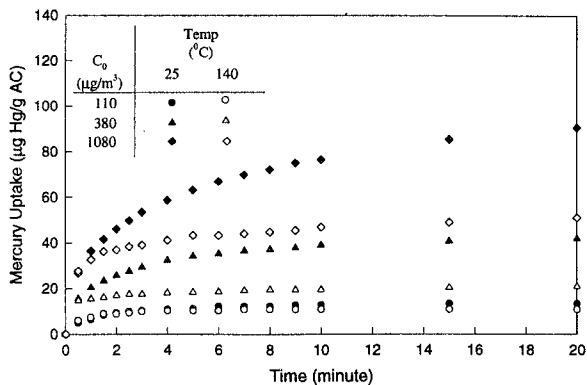


Figure 2. Mercury Uptake by BPL Carbon

### Mercury Adsorption by BPL-S Carbon

In addition to adsorption and diffusion issues with Hg uptake by BPL carbon, Hg uptake by sulfur impregnated carbons also involves the reaction of mercury and sulfur on the carbon surface. Adsorption experiments with BPL-S carbon are shown in Figure 3.

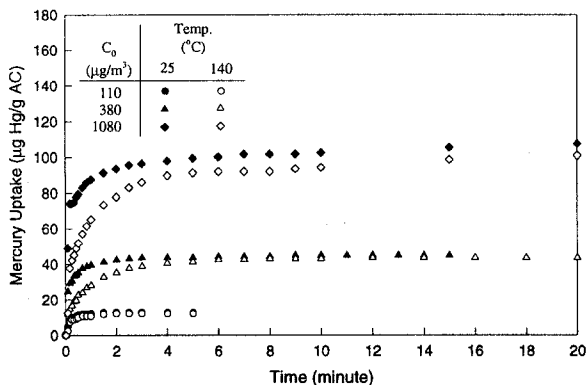


Figure 3. Mercury Uptake by BPL-S Carbon

As shown in Figure 3, the overall Hg uptake for the initial concentration of  $1080 \mu\text{g}/\text{m}^3$  is higher than that observed for  $380 \mu\text{g}/\text{m}^3$ . However, it appears that a dynamic equilibrium is reached at the same time in both experiments. Therefore, the kinetics of Hg uptake at higher initial concentration must have been faster to allow both systems to reach stable levels at the same time.

From the analysis of Hg adsorption data by BPL carbon, it can be concluded that faster kinetics is achieved by increasing the gas temperature. Since Figure 3 shows that the rate of equilibration in the system decreased slightly with an increase in temperature, it can be concluded that the rate of chemical reaction between Hg and S is the rate limiting step for mercury uptake by BPL-S carbon.

### Mercury Adsorption by HGR Carbon

Mercury adsorption experiments with HGR carbon are shown in Figure 4. For both initial concentrations of 380 and  $1080 \mu\text{g}/\text{m}^3$ , the overall mercury uptake

decreased with an increase in temperature because the reaction of mercury with sulfur is exothermic. Since the time required to reach the dynamic capacity did not change at the temperatures of 25 and 140 °C, it can be concluded that the uptake kinetics are slower at higher temperatures than at lower temperatures. This behavior is opposite of what is expected if either diffusion or reaction is the rate limiting step and can be explained by the change in sulfur structure at 140 °C. Since 140 °C is above the melting point of elemental sulfur ( $115.21^\circ\text{C}$ )<sup>(26)</sup>, the weakly bonded sulfur in the macroporous region of an HGR carbon particle will melt and agglomerate in the form of long polymer chains, thereby decreasing the availability of terminal sulfur atoms to react with mercury<sup>(25)</sup>. This suggests that the rate of HgS diffusion into the sulfur mass is the limiting step in the overall process of mercury removal by HGR carbon.

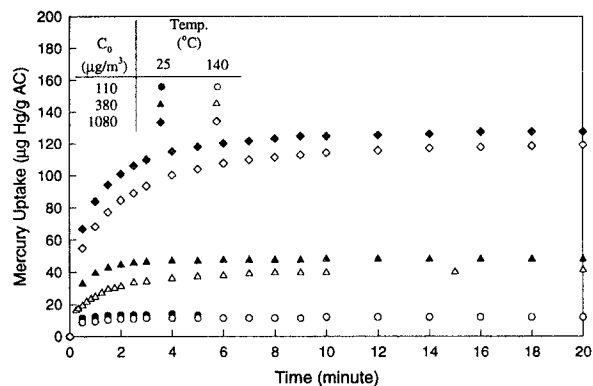


Figure 4. Mercury Uptake by HGR Carbon

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