Development of Novel Activated Carbon-Based Adsorbents for Control of Mercury Emissions From Coal-Fired Power Plants

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Abstract

Objectives

The overall objective of this study is to evaluate pertinent design and operational parameters that would enable successful application of adsorption-based technologies for the reduction of mercury emissions from coal-fired power plants. The first part of the study will evaluate the most suitable impregnate for its ability to enhance the adsorptive capacity of activated carbon for mercury vapor under various process conditions. The second part of the study will evaluate the rate of mercury uptake (adsorption kinetics) by several impregnated activated carbons. Concerned with the ability of the adsorbed mercury to migrate back into the environment once saturated adsorbent is removed from the system, the study will also determine the fate of mercury adsorbed on these impregnated carbons.

Approach and Findings

Novel sulfur-impregnated activated carbons were designed and developed in this study. A commercially available virgin activated carbon (BPL, Calgon Carbon Corporation, Pittsburgh, PA) was used as the starting material. BPL was impregnated with elemental sulfur vapor in an inert atmosphere at different temperatures and sulfur-to-carbon ratios (SCR). The new carbons were named based on the impregnation condition. For example, the adsorbent impregnated with sulfur at 600 °C using the SCR of 4:1 is denoted as BPL-S-4/1-600. Commercially available sulfur impregnated carbon (HGR) was also evaluated for the efficiency of mercury uptake.

Virgin and sulfur-impregnated carbon were studied in a fixed bed reactor. Mercury-laden gas (55 $\mu g/m^3$) was passed through the reactor in a down-flow mode. The system was operated in a continuous mode under constant temperature (140 °C) and 1 atm conditions. An atomic adsorption spectrometer (AAS) was used to monitor the effluent concentration of mercury from the reactor.

Sulfur-impregnated activated carbons manufactured using the novel impregnation procedure had demonstrated much higher efficiency of mercury removal in comparison to BPL and HGR carbons. For example, HGR reached 100-percent mercury breakthrough after only 3 hours, yielding a mercury uptake capacity of 44 μg Hg/g carbon. On the other hand, BPL-S-4/1-250 reached 100-percent mercury breakthrough after 96 hours, resulting in the adsorption capacity of 594 μg Hg/g carbon.

For the BPL-S series, impregnation temperature was one of the most important factors which determined the efficiency of mercury uptake. Higher impregnation temperature resulted in better mercury uptake for the experimental conditions employed in this study. The time to reach 100-percent mercury breakthrough for BPL-S-4/1-250, BPL-S-4/1-400, and BPL-S-4/1-600 was 96, 192, and 408 hours, respectively, and the resulting mercury uptake capacity was 594, 1687, and 2376 μg Hg/g carbon.

SCR was another important parameter for determining the efficiency of the BPL-S series for mercury adsorption. The mercury removal efficiency decreased with a decrease in SCR. For example, the mercury uptake for BPL-S-2/1-600 was 1900 μg Hg/g carbon, while the capacity of BPL-S-1/1-600 was only 1200 μg Hg/g carbon. However, SCR had less impact on mercury uptake as compared to the impact of impregnation temperature.

Sulfur has different allotropes at different temperatures. At higher temperatures, the major forms of sulfur are short chain molecules which have more active sulfur atoms. Thus, a greater portion of active sulfur atoms were attached to the carbon surface if the impregnation process was done at higher temperatures. As a consequence, carbons impregnated at higher temperatures have higher capacity for mercury uptake.

Specific surface area also strongly influenced the performance of these carbons. Higher impregnation temperatures resulted in larger surface areas of BPL-S carbons. Since larger surface area offers more active sites for the reaction between sulfur and mercury, more mercury could be adsorbed onto these carbons.

Tests of pore size distribution revealed another key point for the efficiency of these adsorbents for mercury removal. Carbons prepared at higher temperatures had relatively more larger pores. Therefore, it was much easier for mercury molecules to enter the carbon pores due to the fact that Knudsen diffusion was the predominant step for mass transfer.

Sulfur content was not a crucial factor for mercury removal. The sulfur content of BPL-S-250 was much higher than any other carbon. However, it exhibited the lowest efficiency for mercury uptake.

Highlights of Accomplishments

New types of sulfur-impregnated carbons that exhibit higher mercury removal capacity than commercially available activated carbon (HGR) have been developed in this study. Increase in the impregnation temperature will create more active sulfur atoms per unit surface area of activated carbon. Consequently, more mercury can be captured by these new carbons. Larger specific surface area and larger fraction of bigger pores are important factors for improving the performance of these mercury sorbents.

Acknowledgment

Funding for this work was provided by the U.S. Department of Energy's Federal Energy Technology Center, under Contract DE-FG22-96PC96212 with the University of Pittsburgh. The results described herein were obtained during the period of performance from September 1996 to June 1997.