

Carbon-Based Sorbents For The Removal of Mercury From Flue Gas

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ABSTRACT

A laboratory-scale packed-bed reactor system is used to screen carbon sorbents for their capability to remove elemental mercury from various carrier gases. When the carrier gas is argon, an on-line atomic fluorescence spectrophotometer (AFS), used in a continuous mode, monitors the elemental mercury concentration in the inlet and outlet streams of the packed-bed reactor. The mercury concentration in the reactor inlet gas and the reactor temperature are held constant during a test. For more complex carrier gases, sorbent capacity is determined off-line by using a cold vapor atomic absorption spectrophotometer (CVAAS) or an inductively coupled argon plasma atomic emission spectrophotometer (ICP-AES). The capacities and breakthrough times of several commercially available activated carbons, as well as novel carbon sorbents derived from flyash, were determined as a function of various parameters. The mechanisms of mercury removal by the sorbents are suggested by combining the results of the packed-bed testing with various analytical results.

Activated carbons can remove mercury from flue gas produced by the combustion of coal. However, there are problems associated with the use of activated carbons for mercury removal from flue gas. Since activated carbons are general adsorbents, most of the components of flue gas will adsorb on carbon, with some in competition with mercury. Carbon sorbents operate effectively over a limited temperature range, typically working best at temperatures well below 300°F. The projected annual costs for an activated carbon cleanup process are high, not only because of the high cost of the sorbent, but also because of its poor utilization/selectivity for mercury. Carbon-to-mercury weight ratios of 3,000:1 to 100,000:1 have been projected. In addition, activated carbons can only be regenerated a few times before exhibiting an unacceptably low activity for mercury removal. Therefore, the development of improved activated carbons, as well as novel sorbents, merits further research.

Many of the experiments in this study used a gas feed of 585 ppb elemental mercury in argon. This is dramatically different than the composition of a typical flue gas from a coal-fired utility. Some of the components in a typical flue gas (e.g. acid gases) can adsorb on an activated carbon and could possibly hinder or help the adsorption of mercury on carbon. The ultra-high purity argon carrier gas was selected to maximize the sensitivity of the AFS for elemental mercury. Mixtures of nitrogen and oxygen were also used as a carrier gas, but in these cases, interferences in the AFS detector led to analyzing the sorbent with CVAAS or ICP-AES. Future testing will investigate whether sorbent capacity differences exist with the

aforementioned carrier gases and actual flue gas. Also, the temperatures at which sorbent capacities were determined are 140EF, 280EF, and 350EF. These temperatures were chosen because of their potential relevance to coal-fired utilities. If a sorbent were contacted with the flue gas by injection into the duct work of a coal-fired utility after the air preheater but before the particulate collection device, it would experience temperatures in the range of 350EF to 280EF. If a sorbent was placed downstream of a wet scrubber, it would encounter a temperature near 140EF.

Three classes of carbon sorbents have been examined. These categories are: 1) unpromoted activated carbons, 2) chemically promoted activated carbons, and 3) carbons derived from flyash that were obtained from utilities or from pilot-scale combustion units located on-site at The Federal Energy Technology Center. The unpromoted activated carbons exhibit a small capacity for elemental mercury, probably due to physical adsorption. Carbons chemically promoted with chlorine, iodine, or sulfur exhibit much larger capacities; the mechanism of mercury removal is related to chemisorption and/or chemical reaction. All the carbon sorbents demonstrated much greater capacities at the lower temperatures.

The untreated carbons separated from flyash exhibited a small capacity for the removal of elemental mercury from argon. This is similar to the behavior of the unpromoted commercially available activated carbons. The carbons derived from flyash showed much larger capacities after treatment with chlorine. Also, since the carbons extracted from flyash are not activated and possess a small surface area, an effort was made to increase the surface area by thermal oxidation in air at 750EF.

The results obtained from the packed bed unit require judicious interpretation when attempting to extrapolate relevance to sorbent introduction via duct injection on an industrial size combustor. One unpromoted activated carbon was also studied in the FETC 500-lb/hr pilot-scale combustor unit for the removal of mercury from the flue gas. When introduced at a sorbent-to-mercury ratio of around 5,000 to 1, the unpromoted activated carbon achieved a high level of mercury removal. However, the used activated carbon recovered from the 500-lb/hr unit had mercury levels of less than 300 ppm (0.3 mg Hg/gram). Unpromoted activated carbons sequester elemental mercury via physical adsorption and therefore exhibit small capacities. Nevertheless, duct injection at large sorbent-to-mercury ratios allows them to achieve high levels of mercury removal from flue gas.