



Mercury oxidation and adsorption characteristics of chemically promoted activated carbon sorbents

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ABSTRACT

Previous entrained-flow tests conducted under elemental mercury (Hg^0)-laden air found that significant amounts of oxidized mercury (Hg^{2+}) are not adsorbed onto cupric chloride-impregnated carbon (CuCl_2 -AC) and brominated activated carbon (DARCO Hg-LH), but entrained to the gas phase. In this study, these sorbents were tested in a fixed-bed system and a filter-added entrained-flow system to further investigate Hg^0 oxidation and adsorption characteristics of CuCl_2 -AC and DARCO Hg-LH. These test results suggested that CuCl_2 -AC has different sites available for Hg^0 oxidation and Hg adsorption, and the resultant oxidized mercury generated from the reaction between Hg^0 and CuCl_2 is re-adsorbed at the site of CuCl_2 -AC available for adsorption. The resultant oxidized mercury was also found to be easily re-adsorbed onto CuCl_2 -AC and DARCO Hg-LH in the filter connected to the entrained-flow reactor.

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1. Introduction

A number of laboratory and field studies were conducted to develop economical and efficient sorbents for reducing mercury emissions from coal-fired power plants. Among the sorbents, raw activated carbon has been extensively tested and has demonstrated relatively good performance in mercury emissions control from bituminous coal-burning units [1–4]. Brominated activated carbon has also been suggested to be effective to enhance mercury removal efficiency especially for subbituminous coal or lignite-burning units which produce relatively low concentrations of oxidized mercury in flue gases [5,6]. Several studies are found to test the performance of activated carbons in Hg^0 removal and investigate Hg^0 removal mechanisms. For example, Olson et al. have proposed that raw activated carbon has two or more reaction sites to remove mercury, and Lewis basic sites on activated carbon can bind oxidized forms of mercury (Hg^{2+}) [7,8]. Zeng et al. [9] showed a significant increase in Hg^0 adsorption by impregnating ZnCl_2 onto activated carbon from their fixed-bed tests, and proposed that chlorine (Cl)-contained functional groups may contribute to the chemisorption of Hg^0 . In addition, Ghorishi et al. [10] demonstrated efficient Hg^0 adsorption of a Cl-impregnated activated carbon in their entrained-flow system.

These studies focused on the Hg^0 adsorption of chemically promoted activated carbon sorbents. Therefore, Hg^0 oxidation and adsorption mechanisms by these sorbents are neither well understood through controlled laboratory studies nor clearly demonstrated through experimental studies.

Our previous study [11] examined Hg^0 control capabilities of cupric chloride-impregnated activated carbon sorbents (CuCl_2 -ACs) and brominated activated carbon (DARCO Hg-LH, Norit Americas Inc., Marshall, TX, USA) in an entrained-flow system simulating in-flight mercury capture in coal-fired flue gas. CuCl_2 -ACs demonstrated higher Hg^0 oxidation and similar Hg adsorption capabilities, compared to DARCO Hg-LH. A similar trend of results was found between these sorbents from the tests conducted with the injection of 1500 ppmv sulfur dioxide (SO_2) [11], although SO_2 is known as the most applicable inhibitor for Hg^0 removal by carbon-based sorbents [2,7]. In addition to the Hg^0 removal capability, the previous entrained-flow results showed that significant amounts of oxidized mercury are not adsorbed onto CuCl_2 -ACs and DARCO Hg-LH, but entrained to the gas-phase. Therefore, this study further investigated Hg^0 oxidation and adsorption characteristics of these sorbents. CuCl_2 -AC and DARCO Hg-LH were tested in a fixed-bed system to investigate Hg^0 oxidation characteristics, and in a filter-added entrained-flow system to test the re-adsorption of the effluent oxidized mercury in the filter. In addition, results obtained from the previous entrained-flow tests [11] were statistically analyzed to further understand the mechanisms of Hg^0 oxidation and adsorption onto CuCl_2 -AC. The tests in this study were conducted in an Hg^0 -laden air stream to look into the Hg^0 oxidation and Hg adsorption under the minimum influence of other flue gas components. Since commercial raw activated carbon (DARCO

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Hg, Norit Americas Inc., Marshall, TX, USA) was found to have very low Hg⁰ removal capability under air flow in the fixed-bed and entrained-flow systems, the test results of chemically promoted activated carbon sorbents were discussed in this study.

2. Experimental

2.1. Sorbent preparation

Commercial brominated activated carbon (DARCO Hg-LH) manufactured by Norit Americas Inc. (Marshall, TX, USA) and 6.5% cupric chloride-impregnated activated carbon (C-AC) synthesized in our laboratory were used in this study. 0.5 g cupric chloride dihydrate (99.0 + % CuCl₂·2H₂O, Aldrich Chemical. Co., Milwaukee, WI, USA) was dissolved in 100 mL isopropyl alcohol (99.8%, Pharmco Products Inc., Brookfield, CT, USA). 4.5 g commercial raw activated carbon (DARCO Hg, Norit Americas Inc., Marshall, TX, USA) was then added slowly, and the mixture was kept at room temperature for about 3 h with continuous stirring. The solution was then filtered, and the resulting solid was dried at 100 °C for approximately 2 h to create cupric chloride-impregnated activated carbon. The filtered solution was analyzed for the residual concentrations of copper and chloride in the solution using Atomic Absorption Spectrophotometer (AAAnalyst 300, Perkin Elmer, Waltham, MA, USA) and Ion Chromatography (DX-600, Dionex, Sunnyvale, CA, USA), respectively. The analysis results showed that 6.5%(w) cupric chloride was impregnated onto the activated carbon. In addition, DARCO Hg-LH and C-AC showed similar surface areas and mean particle diameters in our previous analysis. The detailed characterization of these sorbents was included in our previous publication [11].

2.2. Fixed-bed system and experimental procedure

A fixed-bed reactor system was constructed as shown in Fig. 1. The 1.27 cm diameter reactor made of borosilicate glass was placed inside a temperature-controllable convection oven (Stabil-Therm® Electric

Utility Oven, Model OV-500C-2, Blue M Electric Company, New Columbia, PA, USA) at a temperature of 140 °C. The detailed specification of the fixed-bed system can be found in our previous publication [12].

25 mg of each sorbent was premixed with 6 g of silica (Fine granules, 40–100 Mesh, Fisher Scientific Inc., Pittsburgh, PA, USA) and placed in the fixed bed. A total air flow rate of 1 L/min was provided to the fixed bed. An Hg⁰-laden gas stream was generated from elemental mercury permeation tubes (VICI Metronics, Inc., Poughkeepsie, NY, USA), and the flow rate was maintained at 100 mL/min using a mass flow controller. An additional air flow of 900 mL/min was injected to the fixed bed. The total flow rate was also monitored using a bubble flow meter. The outlet mercury speciation was conducted by using the Ontario Hydro Method [13]. A 1 M KCl solution and 5% (v/v) HNO₃/10% (v/v) H₂O₂ solution were used to capture oxidized mercury. The outlet Hg⁰ concentration was also observed by an online mercury analyzer (UV-1201S with mercury analysis kit, Shimadzu Corp., Columbia, MD, USA), and an acidified KMnO₄ solution was used to capture elemental mercury from the effluent gas. After each test, the spent sorbent was collected and analyzed for the amount of mercury adsorption by following the digestion procedures described in the Ontario Hydro Method. Although Hg adsorption onto each sorbent was found to be essentially complete based on these analyses during the fixed-bed test, the test was continued to investigate Hg⁰ oxidation characteristics of each sorbent. Therefore, these fixed-bed results may not be applicable to mercury capture in coal-fired flue gas.

2.3. Filter-added entrained-flow system and experimental procedure

In the previous study [11], significant amounts of effluent oxidized mercury were found from the tests of DARCO Hg-LH and CuCl₂-ACs conducted in an 8 m-long 2.5 cm diameter entrained-flow reactor. A 10-cm diameter filter was connected to the end of the entrained-flow reactor to test the re-adsorption of the effluent oxidized mercury in the filter and to simulate mercury capture in the ductwork and fabric filter of coal-fired utilities. The filter holder was

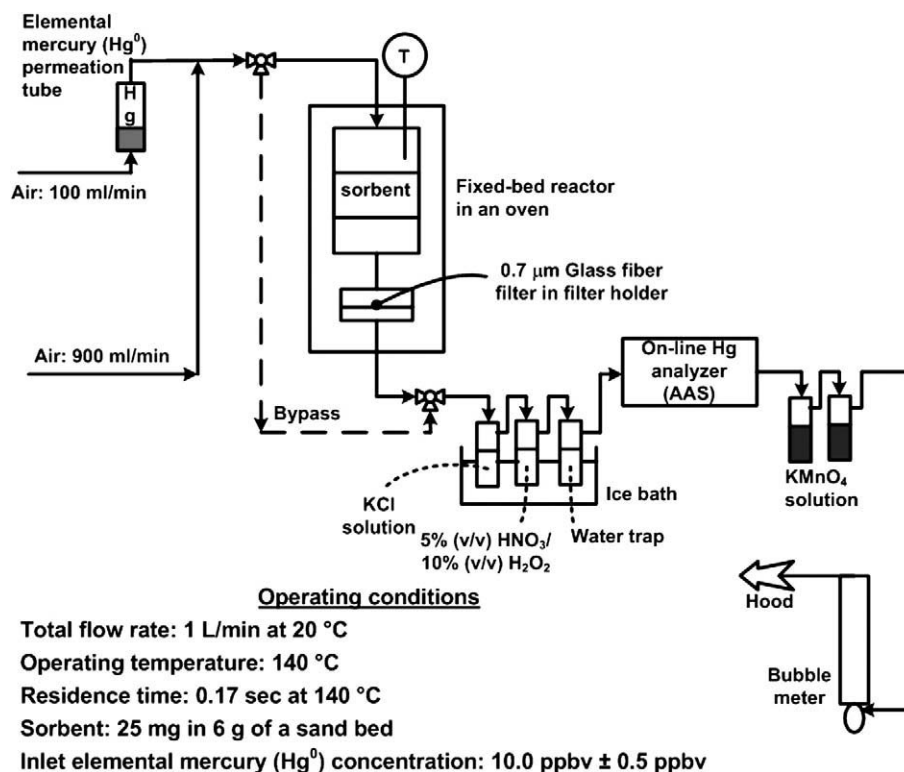


Fig. 1. Schematic and operating conditions of the fixed-bed system.

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