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Removal of elemental mercury from flue gas by recyclable $CuCl_2$ modified magnetospheres from fly ash. Part 4. Performance of sorbent injection in an entrained flow reactor system

Jianping Yang[a,b], Yongchun Zhao[b,*], Xin Guo[b], Hailong Li[a], Junying Zhang[b], Chuguang Zheng[b]

[a] School of Energy Science and Engineering, Central South University, Changsha 410083, China
[b] State Key Laboratory of Coal Combustion, School of Energy and Power Engineering, Huazhong University of Science and Technology, Wuhan 430074, China

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ABSTRACT

Experimental studies on Hg^0 removal by $CuCl_2$ modified magnetospheres (Cu_6–MF) injection were performed in an entrained flow reactor system. The factors affecting the Hg^0 removal efficiency were investigated, including the initial Hg^0 concentration, sorbent concentration in flue gas, sorbent particle size, sorbent residence time in flue gas, and flue gas temperature. The results indicated that, as the raise of initial Hg^0 concentration, the Hg^0 removal efficiency increased. The Hg^0 removal efficiency also increased with the increase of sorbent residence time and sorbent concentration in flue gas. When the residence time was 1.61 s and the sorbent concentration was 1.09 g/m^3, the Hg^0 removal efficiency could reach to 80.6%. However, with the further increase of residence time and sorbent concentration, the Hg^0 removal efficiency did not increase continuously. The smaller particle size was favorable for the Hg^0 removal. In the industrial application, the particle size could be selected as 45–74 μm when comprehensively considering the Hg^0 removal efficiency, the cost of sorbent preparation, and other factors. The acid flue gases have negligible impacts on Hg^0 removal, while moisture plays a slightly negative role in Hg^0 removal under SFG atmosphere. With the superior Hg^0 removal performance, the Cu_6–MF sorbent may presents great potentials for Hg^0 removal in coal-fired power plants.

1. Introduction

Mercury (Hg) emission and pollution has attracted significant attention in whole world due to its high toxicity in human health and the environment [1,2]. At 16th August 2017, the "Minamata Convention", which aims to protect human health and the environment from anthropogenic Hg emissions, has come into force for all the signatories. Although many countries including United States and China have issued national Hg emission standards before December 2011, it can be predicted that more restrict Hg emission limits will be announced to realize the goal of "Minamata Convention".

Coal-fired power plants are one of the largest anthropogenic Hg emission sources [1]. In recent years, a variety of Hg removal technologies have been developed for Hg removal from coal-fired power plants. The injection of sorbent upstream of either electrostatic precipitator (ESP) or fabric filters (FF) in coal-fired power plants is one of the most promising control technologies. Nowadays, various Hg sorbents, including carbon-based sorbents [3–13], fly ash [14–17], natural mineral sorbents [18], noble metals [19–23], metal oxides [24–37], sulfur compounds [38,39], and some novel materials [40,41] have been developed. Of these different potential sorbents, activated carbon (AC) is one of the most commercialized sorbent for Hg^0 removal. Although it is demonstrated that AC possesses high Hg^0 adsorption capacity, other issues should be considered, such as the high cost of sorbent preparation. Particularly, the raw AC is generally modified by sulfur or halogens to improve the Hg^0 adsorption capacity, which will results in the increase of sorbent preparation cost. In addition, the carbon content of fly ash will be increased by AC injection, depreciating its reuse as a raw material for concrete. Thus, it is urgently needed to develop lost-cost non-carbon sorbent for the alternative of AC.

In our previous studies [42–44], an effective and low-cost recyclable $CuCl_2$ modified magnetospheres (Cu_x–MF) sorbent was developed. In these series studies, the Hg^0 removal performance and the involved reaction mechanism were investigated based on a fixed-bed reactor system. It was demonstrated that the optimal sample of Cu_6–MF obtained above 90% of Hg^0 removal efficiency under simulated flue gas (SFG) atmosphere, and the acid flue gases (SO_2, NO, and HCl) played negligible negative roles in Hg^0 removal. The fixed-bed tests can provide useful information including Hg breakthrough curve and adsorption equilibrium parameters. However, in the actual power plants, the

* Corresponding author.
E-mail address: yczhao@hust.edu.cn (Y. Zhao).

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sorbent is generally injected into duct by injector for Hg^0 removal. Besides the Hg^0 adsorption capacity, the Hg^0 removal performance will be affected by the gas-particle two-phase flow characteristics and the gas-phase mass transfer. In this way, the fixed-bed tests cannot reflect the Hg^0 removal performance of sorbent injection. Even though it can obtain the most reliable information of Hg^0 removal performance at realistic conditions through field tests, they have notable disadvantages like high cost and operating difficulties. In this way, to facilitate the industrial application of Cu_6–MF in in-duct Hg^0 removal, a study on Hg^0 removal performance in an laboratory-scale entrained flow reactor system is of considerable importance.

In this work, experimental studies on in-duct Hg^0 removal by Cu_6–MF injection were performed in an entrained flow reactor system. The factors affecting the Hg^0 removal efficiency were investigated, including initial Hg^0 concentration, sorbent concentration, particle size, residence time, and flue gas temperature. The effects of acid flue gas components (SO_2, NO, HCl) and moisture on Hg^0 removal performance were studied. In addition, the Hg^0 adsorption and oxidation performances of sorbent injection were also identified.

2. Materials and methods

2.1. Sample preparation and characterization

In this work, the sample of Cu_6–MF, which presented optimal Hg^0 removal performance in our previous study, was used to evaluate the in-duct Hg removal performance. The sorbent was prepared by an incipient wetness method, which was described in Part 1 of this series study [42]. Briefly, fly ash was collected from the electrostatic precipitator hoppers of a typical pulverized coal-fired power plant in China, and the magnetospheres were recovered from fly ash by wet magnetic separation. After that, the magnetospheres were ground and size fractionated by screening (< 45 μm, 45–74 μm, 74–125 μm). Subsequently, the magnetospheres were impregnated by a cupric chloride ($CuCl_2{\cdot}2H_2O$) solution at room temperature for 2 h. The Cu loading value of the catalyst was 6%. After the impregnation process, the sample was firstly dried at 30 °C for 12 h and then dried at 110 °C for 8 h. The physical-chemical characteristics of prepared samples were shown in Part 1 of the series [42].

2.2. Experimental apparatus and procedures

The in-duct Hg removal performance was investigated in an entrained flow reactor apparatus, as shown in Fig. 1. The experimental apparatus consisted of the following parts: Hg^0 permeation device, simulated flue gas feed devices, gas heater, gas premix chamber, sorbent injection device, duct (Hg removal reaction system), sorbent collection device (cyclones), gas purification devices (filters), on-line Hg analyzer. The duct of simulated flue gas was 20 m long and 16 mm inside diameter, which was made of stainless steel with Teflon lining on internal wall surface. The duct was heated by electric heating tape to obtain the desired flue gas temperature. Five thermocouples were installed along the length of the duct uniformly to monitor the flue gas temperature. To investigate the effect of sorbent residence time in flue gas on Hg^0 removal efficiency, four sampling sites were located on the duct to measure the Hg concentration.

The total flow rate of simulated flue gas was 58 L/min, with the gas speed of 9 m/s, which was similar to that in ESP inlet of actual power plants. The total flow consisted of three parts. The first part of flow was pure N_2 from a cylinder, which was used as carrier gas of Hg^0 vapor, keeping the flow rate of 3 L/min. The Hg^0 vapor was generated from a Hg^0 permeation tube (VICI, Metronics Inc., Santa Clara, CA). To investigate the effect of initial Hg^0 concentration on Hg removal efficiency, the concentration was set as 4.7 μg/m^3, 9.5 μg/m^3, 16.4 μg/m^3, respectively. The second part of flow was used to carry the sorbent into duct, with a flow rate of 15 L/min, which was generated from an air compressor. The sorbent was fed into the duct inlet from the sealed chamber by using a micro-screw feeder and carried into the duct by an injector with the assistance of purified air. The sorbent concentration injected into flue gas was controlled by adjusting the screw rotation speed of speed controller. The third part was generated from the air compressor as well, which mixed with the other two parts after passed through the flue gas heater. This part of flow acted as the main component of the simulated flue gas and the flow rate was 40 L/min. Moreover, acid flue gas components (1200 ppm SO_2, 300 ppm NO and 10 ppm HCl) and 5% water vapor were also introduced into the system together with the third part of flow. The sorbent residence time in flue gas was 0.55 s, 1.12 s, 1.61 s, and 2.24 s at sampling site #1, #2, #3, and #4, respectively. The flue gas temperature was set as 100 °C, 125 °C, 150 °C, respectively, with an intention to investigate the effect of flue gas temperature on Hg^0 removal performance. In order to increase the accuracy of sorbent injection rate, the sorbent was mixed with neutral Al_2O_3 with same particle size, and the mass ratio of sorbent and neutral Al_2O_3 was 1:9. The injection rate of sorbent was 0.22, 0.44, 0.89, and 1.09 g/m^3, respectively. Part of flue gas was extracted at each sampling site to measure the Hg^0 concentration in flue gas, and the flow

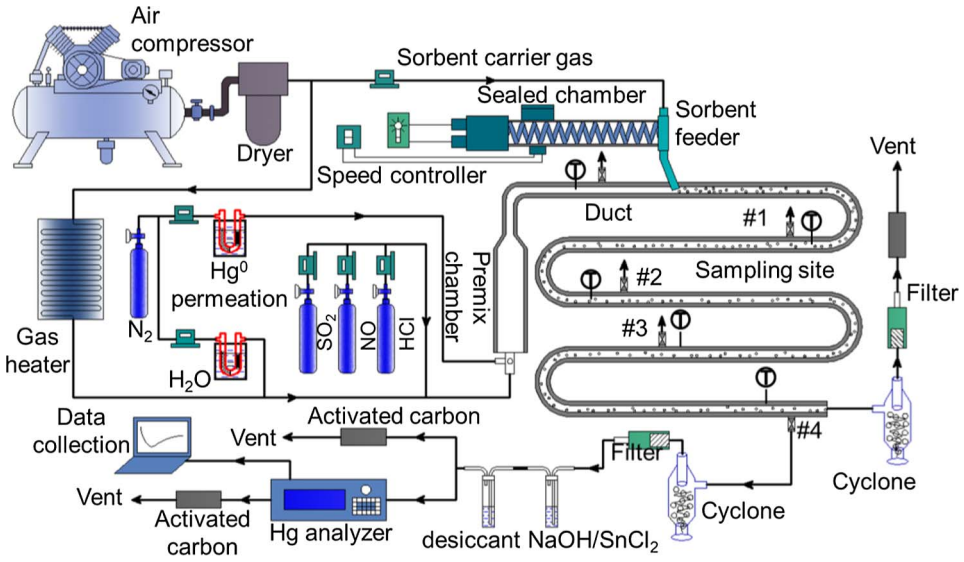

Fig. 1. Schematic diagram of entrained flow reactor apparatus.