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Research article

Modeling and experimental studies of in-duct mercury capture by activated carbon injection in an entrained flow reactor

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ABSTRACT

A new mathematical model was proposed to predict in-duct mercury capture efficiency and determine the effect of sorbent properties on mercury capture by sorbent injection. The model was based on external film mass transfer assumption and included mass balance and adsorption isotherm. Adsorption parameters of the sorbent were determined by the fixed-bed mercury adsorption experiments and other modeling parameters were estimated from literatures. The model verification was conducted by comparing with the experimental results of mercury capture by raw and bromine modified activated carbon (R-AC and AC-Br) injection in an entrained flow reactor. The results show that there is a reasonable agreement between the modeling prediction and the experimental results, which demonstrates that this model can provide a rational prediction results and be used to estimate sorbent consumption cost. In both the experimental and modeling cases, mercury capture efficiencies are improved with smaller sorbent particle size, longer sorbent residence time and larger sorbent concentration. The in-duct mercury capture rate of AC-Br was improved significantly compared to that of R-AC. The simulation of in-duct mercury capture by sorbent injection demands taking into account the additional mercury capture by deposited activated carbon on the duct wall. The model parameters, including sorbent concentration, particle size, equilibrium constant *K*, external film mass transfer coefficient and residence time have important effects on the in-duct mercury capture rate by sorbent injection.

1. Introduction

Mercury emission from coal-fired power plants has attracted worldwide concern because of its high toxicity, volatility, long-distance migration, and bioaccumulation in the atmosphere [1–4]. It is generally agreed that the mercury in coal will be vaporized and converted to elemental mercury (Hg^0) completely in combustion zone [5,6]. Part of Hg^0 discharged from the boiler will transform to oxidized mercury (Hg^{2+}) and particle-bound mercury (Hg^p) as flue gas temperature decreases [7]. Hg^{2+} is soluble in water and can be easily removed by the wet flue gas desulfurization (WFGD) facilities. Hg^p can be captured with fly ash in typical particle control devices (PCDs) such as electrostatic precipitators (ESPs) and fabric filters (FFs) [8–10]. However, it is difficult to remove Hg^0 by WFGD or PCDs because of the high volatility and insolubility [11,12]. The emitted Hg^0 in the atmosphere will form methyl mercury, which is seriously harmful to human beings [13,14]. An effective way of preventing Hg^0 from being released into the atmosphere is to transform it into Hg^{2+} and/or Hg^p which are easier to be captured.

Sorbent injection technology is considered to be the most promising and effective method for controlling mercury emission from the power plants [15,16]. Sorbent evaluation has been widely conducted in fixed-bed [17,18], entrained flow reactor [19,20] and field tests [21,22], in order to develop the sorbents with low cost and high mercury capture capability. Fixed-bed tests can provide mercury breakthrough curve and adsorption equilibrium parameters, as well as simulate mercury capture in fabric filters, but it cannot reflect the in-duct mercury capture performance of sorbent. Field tests can generate the performance information of sorbents at realistic conditions, but they have the shortages of high cost and operational difficulties, discouraging the obtaining of many important parameters. Therefore, entrained flow tests have been widely utilized to evaluate in-duct mercury capture performance of different sorbents and to obtain the optimal operating parameters before field test [19,20,23,24].

To predict mercury capture efficiency, identify optimal operation conditions, and estimate sorbent consumption cost of sorbent injection, an easy-to-use and accurate model is needed. However, the models developed so far to predict sorbent injection for mercury capture are limited. Most existing models are based on the following fundamental theory to describe the process: three steps are assumed, and the steps include external film mass transfer, intraparticle diffusion and adsorption at activate sites [25–32]. Meserole et al. [25] developed a theoretical model to predict mercury removal by sorbent injection. The model considered two steps of gas-phase mass transfer and surface reaction. Flora et al. [26,27] established a two-stage model describing Hg removal in

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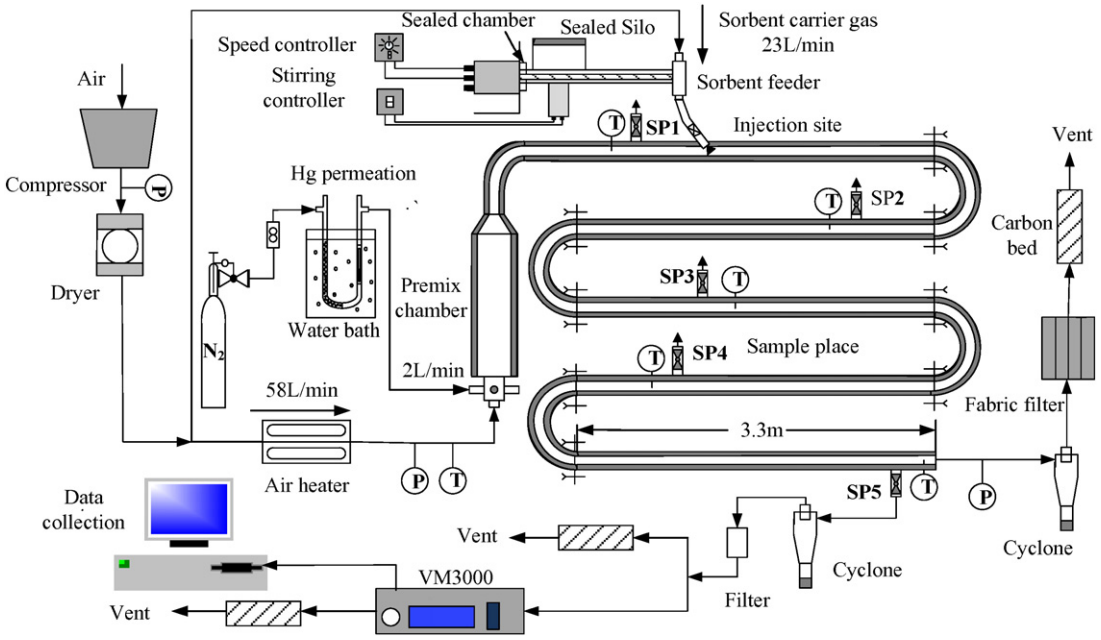

Fig. 1. Schematic diagram of entrained flow reactor apparatus.

the duct and in the fabric filter using the pore diffusion model (PDM). In their model, a plug flow system was used to simulate mercury removal in the duct, while a packed-bed approach was used to simulate mercury removal in the fabric filter. Scala et al. [28–30] combined the process of external mass transfer and intraparticle diffusion for predicting mercury capture in incinerator flue gas. Zhou et al. [31] developed a CFD model to predict mercury capture rate and evaluate the lance design. The model described the external film mass transport, pore diffusion, surface adsorption and desorption process. Zhao et al. [32] proposed a simplified model, and it was based on the combination of external mass transfer, intraparticle diffusion and isothermal adsorption. All these models consider the complicated process of external film mass transfer and intraparticle diffusion or surface reaction. However, the intraparticle diffusion is a very complex process including surface diffusion, molecular diffusion and Knudsen diffusion, which significantly increase the equation numbers and model parameters, leading to the increase of computational complexity and difficulty in model solution as well as the difficulties in obtaining accurate prediction results.

In our previous study [33], we found that mercury adsorption on sorbent could be divided into two stages, including surface adsorption and intraparticle diffusion adsorption. In the initial stage of adsorption, the surface adsorption was the main adsorption form. The residence time of sorbent staying in duct was less than 2 s. In such a short time, the adsorption mainly occurred on the sorbent surface and intraparticle diffusion adsorption hardly occurred. The research [34] by Chen et al. showed that under certain conditions, in-duct mercury capture by carbon injection was controlled by external film mass transfer process, and intraparticle diffusion was not important.

Therefore, in order to simplify in-duct mercury capture process, we developed a new prediction model that included only external film mass transfer process. The model would be used to predict in-duct mercury capture efficiency and determine the effect of sorbent properties on mercury capture by sorbent injection. The model verification was conducted by comparing with the experimental results of in-duct mercury capture by raw and bromine modified activated carbon (R-AC and AC-Br, respectively) injection in an entrained flow reactor. Effects of the model parameters on mercury capture efficiency were also evaluated.

2. Experimental

2.1. Sorbents preparation and characterizations

The sorbents used in the modeling and experiments were R-AC and AC-Br. The AC-Br was prepared by impregnating R-AC sample with 1% (mass ratio) NH_4Br solution. After being stirred for 12 h, the sorbent particles were filtered out and dried in an oven at 45 °C. N_2 adsorption and X-ray energy-dispersive spectroscopy (EDX) characterizations

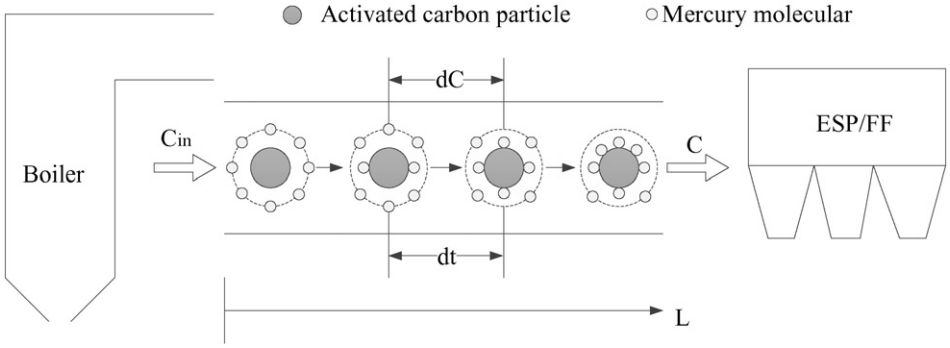

Fig. 2. Schematic of in-duct mercury capture by sorbent injection.