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Research article

Study on the effects of carrier and modifier on mercury adsorption behavior over halides modified sorbents using temperature programmed desorption method

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ABSTRACT

Halides modified sorbents injection equipped with dust removal units is considered as the most promising technology for elemental mercury (Hg^0) removal from coal-fired flue gas. However, Hg^0 adsorption behavior over different halides modified sorbents remains controversial. In this study, the effects of carriers (activated carbon and neutral Al_2O_3) and modifiers (NaCl, NaBr, $CuCl_2$, and $CuBr_2$) on the behavior of Hg^0 adsorption over modified sorbents were investigated using temperature programmed desorption (TPD) method. Both mercury adsorption experiments and TPD experiments were conducted in the bench-scale fixed bed. The results indicated that $CuBr_2$-modified activated carbon (AC) showed the best mercury removal performance due to the lower bond energy of $CuBr_2$. The decomposition temperature of mercury compounds generated on AC was different from that on neutral Al_2O_3. Further analysis indicated that the modifiers with lower bond energy could release halogens during modification. The released halogens could react with AC to form active C—Cl or C—Br group. Moreover, the lower bond energy of modifiers made it easier for halogens to release. But Al_2O_3 carrier could not react with modifiers to form active group during modification. The Hg^0 adsorption process over modified Al_2O_3 could be explained by Langmuir-Hinshelwood mechanism.

1. Introduction

Mercury has strong toxicity to animals and humans through food-chain [1, 2]. According to statistics, coal-fired power plants have become one of the biggest anthropogenic sources of mercury emissions in the world [3–6]. Mercury has three basic chemical forms in coal-fired flue gas: particulate mercury (Hg^P), oxidized mercury (Hg^{2+}) and elemental mercury (Hg^0) [7–10]. Hg^P can be captured by dust control units and Hg^{2+} can be removed by wet flue gas desulfurization due to its good water-solubility [11–13]. However, Hg^0 is insufficiently removed by the existing air pollution control devices because of its high volatility and low solubility in water [14–16]. Therefore, great efforts have been focused on the removal of Hg^0 from coal combustion flue gas. The general method for controlling mercury emission fall into two categories: (1) Hg^0 is converted into Hg^P through sorbents injection [17–20] and (2) Hg^0 is oxidized into Hg^{2+} using strong oxidants or catalysts [21–24].

Among them, halides modified sorbents injection equipped with dust removal units is considered as the most promising technology for Hg^0 removal from flue gas [25–29]. The mercury removal efficiency of sorbents varies from the kind of activated carbon, flue gas composition, flue gas temperature, etc. In coal-fired power plants, the used sorbents after mercury adsorption are often processed together with fly ash. An inappropriate method for disposing fly ash may cause the secondary release of mercury [30]. Thus it is necessary to identify the mercury species on the used sorbents. This could provide guidance for inhibiting mercury emission during fly ash disposal. However, the amount of mercury adsorbed on sorbents was too small to reach the detection limits of measuring instruments. Temperature programmed desorption (TPD) method has been widely adopted to identify mercury species in solids due to its low detection limit and high accuracy [31–33]. TPD method is based on the fact that mercury compounds with different stability decomposed at different temperatures. In this case, mercury species can be judged form the decomposition temperatures. Lopez-Anton et al. have successfully identified and quantified mercury species in fly ash via TPD analysis [34]. Wu et al. used TPD to investigate the reactivity of HgO pretreated by HCl and SO_2 [25]. In our previous studies [35, 36], TPD method was used to identify mercury occurrence in coal and Hg^0 adsorption mechanism over AC's oxygen functional groups. Although TPD is an effective method for identifying the

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