



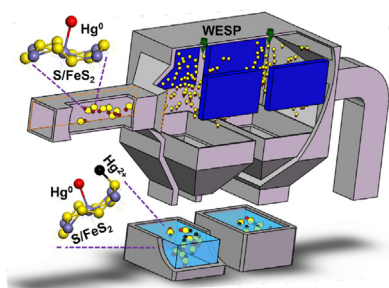
Full Length Article

Sulfur abundant S/FeS₂ for efficient removal of mercury from coal-fired power plants

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GRAPHICAL ABSTRACT



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ABSTRACT

Sulfur abundant S/FeS₂ prepared by a hydrothermal method was employed for removing elemental mercury (Hg⁰) from coal-fired flue gas at low temperature. The S/FeS₂ exhibited optimal Hg⁰ adsorption performance at 80 °C, which matched the temperature window between the flue gas desulfurization (FGD) and wet electrostatic precipitator (WESP) systems. The adverse effects of H₂O and SO₂ on Hg⁰ adsorption were slight. The Hg⁰ adsorption capacity of S/FeS₂ was up to 2732 µg/g when achieved 50% breakthrough threshold. Both elemental sulfur (S) and FeS₂ in the S/FeS₂ contributed to the excellent Hg⁰ adsorption capacity. The mercury leaching tests show that only 0.00076% mercury adsorbed on the S/FeS₂ was leached out. The mercury concentration in leachate was 0.694 µg·L⁻¹, which was much lower than that for a commercial AC (1.214 µg·L⁻¹). Furthermore, the S/FeS₂ presented about 95% oxidized mercury (Hg²⁺) adsorption efficiency from WESP effluent. The Hg²⁺ concentration could decreased rapidly from 50 µg·L⁻¹ to below 2.5 µg·L⁻¹ in 30 min, which was much lower than the World Health Organization (WHO) guideline (6 µg·L⁻¹). With these advantages, S/FeS₂ appears to be a promising material for co-beneficial gaseous Hg⁰ and aqueous Hg²⁺ removal from power plants by injecting upstream of a WESP system.

1. Introduction

The emission and pollution of mercury has raised great attention in the worldwide because of its extreme toxicity, persistence, and bioaccumulation [1,2]. “Minamata Convention on Mercury”, which aims to protect human health and the environment from the adverse effect of

anthropogenic mercury emissions, has come into force in August 16th 2017. Coal combustion is one of the most significant anthropogenic mercury emission sources [2]. To meet the global treaty and regional emission standards, various technologies for removing mercury from flue gas, including adsorption [3–11] and catalytic oxidation [12–18], have been developed in recent years.

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Activated carbon injection (ACI) upstream of a particulate control device (PCD) is regarded as one of the most promising approaches for removing mercury from power plants [19]. However, a high C/Hg ratio of 10,000–100,000 wt./wt. is required to obtain 90% mercury removal efficiency [20]. Hence, the high operating cost impeded this approach's industrial application. The mercury-laden AC is generally captured along with fly ashes by a PCD, the ultimate fate of which is mostly dumped in landfill or used as the raw concrete manufacturing material with fly ashes. The mercury adsorbed on AC might be leached out under prolonged landfill or re-emitted during fly ash utilization [21]. Therefore, it is essential to develop effective economic and ecofriendly alternatives to AC for mercury removal from power plants.

Sulfide minerals with abundant surface sulfur, for which mercury has a high binding affinity, exhibited great potential for mercury adsorption [6,22–26]. However, the sulfide mineral would encounter the poison of high concentration SO_2 and a small amount of NO_x exist in flue gas if applied for mercury adsorption by injecting upstream of a PCD system [22,27]. Meanwhile, the mercury-laden sulfide minerals still face the risk of mercury leaching under prolonged landfill or re-emission during the utilization of fly ash [21]. In recent years, wet electrostatic precipitators (WESPs) are equipped downstream of the flue gas desulfurization (FGD) system to capture ultrafine particles and aerosols. Thus, a promising strategy is to inject the sulfide minerals upstream of a WESP system. In this way, the detrimental effect of SO_2 and NO_x on Hg^0 adsorption is minimized, since most SO_2 and water-soluble NO_x was removed by FGD. Meanwhile, because > 99% fly ashes were removed by PCD, the amount of particulates collected by WESP is much smaller, which makes it more practicable to dispose the mercury-laden sulfide minerals. More attractive, mercury ion (Hg^{2+}) in aqueous solutions is a soft Lewis acid, it has a strong binding affinity for soft Lewis base like reduced-S ligand in sulfide minerals [28–33]. Pyrite (FeS_2), the most abundant sulfide minerals in nature, was a commonly available and inexpensive material for immobilizing Hg^{2+} in wastewater [28–32]. The FeS_2 can react with Hg^{2+} to form hardly soluble HgS , with a solubility product constant (K_{sp}) of 4×10^{-54} [31]. With this advantage, it is promising to inject FeS_2 upstream of a WESP system for capturing gaseous Hg^0 from flue gas and co-beneficially immobilizing Hg^{2+} in WESP effluent. However, to the best of our knowledge, the co-beneficial Hg^0 and Hg^{2+} adsorption over FeS_2 , especially the sulfur abundant FeS_2 , has not yet been reported.

In this work, sulfur abundant S/ FeS_2 was synthesized by a hydrothermal method and employed for capturing Hg^0 at low temperature. The Hg^0 adsorption performances of S/ FeS_2 and other sorbents including commercial AC were compared. The mechanism responsible for the excellent Hg^0 adsorption capacity of S/ FeS_2 was investigated. Furthermore, as S/ FeS_2 was designed to be applied for Hg^0 adsorption by injecting upstream of a WESP, the mercury leachability of mercury-laden S/ FeS_2 as well as the co-beneficial Hg^{2+} removal capacity from WESP effluent were also studied.

2. Experimental section

2.1. Sample preparation

The sample was synthesized by a hydrothermal method [34]. In a typical procedure, 0.05 mol ferrous sulfate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), 0.05 mol sodium thiosulfate ($\text{Na}_2\text{S}_2\text{O}_3 \cdot 5\text{H}_2\text{O}$) and 0.025 mol sublimation of sulfur (S) were dissolved in 50 ml double-distilled water and stirred for 30 min. Then the aqueous solution was transferred into a 100 ml Teflon-lined reactor and kept at 200 °C for 24 h. After cooling naturally to room temperature, the sample was centrifuged and dried in vacuum at 60 °C for 3 h. Finally, the sample was ground and sieved to 60–80 mesh, and the obtained sample was denoted as S/ FeS_2 . A commercial AC used for mercury removal in power plant was purchased from Calgon Carbon Corporation and used for Hg^0 removal tests as comparison.

2.2. Sample characterization

The Brunauer-Emmett-Teller (BET) specific surface area of the S/ FeS_2 was measured by a BET analyzer (ASAP-2020, Micromeritics). Before the BET measurement, the sample was firstly degassed at 60 °C in vacuum for 12 h so as to remove atmospheric gases. The phase structure of the S/ FeS_2 was determined by a X-ray diffractometer (XRD, SIMENS D500 Bruker) operating at 40 kV and 40 mA using a $\text{Cu K}\alpha$ radiation in the range of 5–90° (2 θ). The thermal stabilities of the samples were investigated by thermogravimetric analysis (TG, SDT Q600 V20.5 Build 15). The carrier gas for TG analysis was pure argon (Ar), with a flow rate of 50 ml·min^{−1}. The sample was heated from 30 to 500 °C at a controlling heating rate of 5 °C·min^{−1}. The chemistry states of S, Fe and Hg on the S/ FeS_2 surface were identified by a X-ray photoelectron spectroscopy (XPS, Escalab 250Xi, Thermo Fisher Scientific). The surface atomic concentrations of each elements were calculated by the XPS spectra. The C 1s binding energy value of 284.8 eV was taken as the reference for correcting the observed spectra.

2.3. Hg^0 adsorption activity measurements

The Hg^0 adsorption performances of samples were investigated using a bench-scale reaction system, which was described in our previous study in details [6]. The simulated flue gas was a mixture of N_2 , 5% O_2 , 50–150 ppm SO_2 , 25–75 ppm NO , 4–12% H_2O , and 68 (± 1) $\mu\text{g}/\text{m}^3$ Hg^0 , in which the total flow rate was 1 L·min^{−1}. Gaseous Hg^0 was provided by a Hg^0 permeation tube (VICI Metronics), which was placed in a water bath and kept at 40 °C to obtain a stable Hg^0 vapor source. The generated Hg^0 vapor was introduced into the system by pure N_2 with a flow rate of 0.3 L·min^{−1}. The reactor inlet and outlet Hg^0 concentrations were monitored by an online mercury analyzer (VM3000, Mercury Instruments, Inc.). The other flue gases (N_2 , O_2 , SO_2 and NO) were supplied by cylinders. Concentrations of flue gases were controlled by mass flow controllers. Water vapor was generated by heating an impinger containing H_2O at 70 °C. Pure N_2 with a flow rate of 0.3 L·min^{−1}, serving as a carrier gas, delivered water vapor into the experimental system. The sorbents were placed in a borosilicate glass reactor, with an inner diameter of 10 mm and a length of 550 mm. The reactor was put in a temperature-controlled tubular furnace to kept at a certain temperature. A trap contained AC was located at the experimental system outlet to purge the exhaust gas.

Four sets of experiments were conducted. The experimental conditions are summarized in Table 1. In Set I experiments, the influences of reaction temperature (40–100 °C) on Hg^0 adsorption over S/ FeS_2 were studied. Set II experiments were performed to investigate the Hg^0 adsorption performances of S/ FeS_2 at different gas hourly space velocity (GHSV). In Set III experiments, the influences of H_2O , SO_2 and NO concentrations on Hg^0 adsorption over S/ FeS_2 were investigated. The experiments in Set IV were conducted to compare the Hg^0 adsorption performances of S/ FeS_2 and commercial AC. Before each test, the gas flow bypassed the reactor until the Hg^0 concentration was stable for 30 min, which was denoted as the inlet Hg^0 concentration (C_{in}). Then, the gas flow passed the reactor and the outlet Hg^0 concentration (C_{out}) was recorded. The Hg^0 adsorption efficiency and capacity were calculated by Eqs. (1) and (2), respectively.

$$\eta = \frac{\int_{t_1}^{t_2} (C_{\text{in}} - C_{\text{out}}) \times f \times dt}{\int_{t_1}^{t_2} C_{\text{in}} \times f \times dt} \times 100\% \quad (1)$$

$$C = \frac{1}{m} \int_{t_1}^{t_2} (C_{\text{in}} - C_{\text{out}}) \times f \times dt \quad (2)$$

where η is Hg^0 adsorption efficiency, C is Hg^0 adsorption capacity ($\mu\text{g Hg}/\text{g sorbent}$), m is the mass of sorbent (g), f is the gas flow rate (m^3/h), and t is the adsorption time (h).

To identify the mercury species on the S/ FeS_2 after mercury

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