Efficient Removal of Elemental Mercury from Coal-Fired Flue Gas
over Sulfur-Containing Sorbent at Low Temperatures

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ABSTRACT: In the work, sulfur-containing sorbents were employed to remove elemental mercury (Hg⁰) from coal-fired flue gas. The work used the thermogravimetric analysis, Brunauer–Emmett–Teller method, scanning electron microscopy with energy-dispersive spectroscopy, X-ray diffraction, and X-ray photoelectron spectroscopy to characterize the physicochemical properties of the sorbents. The Hg⁰ removal performance of these used sorbents from the simulated coal-fired flue gas was evaluated by a bench-scale fixed-bed reactor. The results indicated that a generous amount of elemental sulfur covered the surface and pore structure of the used sorbent. With the rise of H₂S selective oxidation temperature, both the sulfur content and specific surface area decreased rapidly. Used-Fe/SC120 could achieve the mercury removal efficiency of above 90% at 90 °C. The high temperature was not conducive to the mercury capture due to the release of surface elemental sulfur. The presence of O₂ and SO₂ inhibited Hg²⁺ removal in different degrees because of the decreased active sulfur sites and competitive adsorption. Meanwhile, NO promoted the Hg⁰ removal efficiency by enhancing the Hg⁰ oxidation. The further analysis showed that the surface elemental sulfur was vital to capture the Hg⁰ from coal-fired flue gas, which reacted with Hg⁰ to form HgS.

1. INTRODUCTION
Mercury is one of the most considerably concerned pollutants in the environment because of its persistence, toxicity, and bioaccumulation in the ecosystem. The anthropogenic mercury sources emitted to the atmosphere environment have been investigated, considering the coal-fired power plant as the main anthropogenic mercury emission source. Generally, there are the three forms of mercury in coal-fired flue gas: particulate mercury (Hg⁰), oxidized mercury (Hg²⁺), and elemental mercury (Hg⁰). The Hg⁰ and Hg²⁺ can be easily removed by existing convention air pollution control units, such as wet flue gas desulfurization, fabric filters, or electrostatic precipitators. However, the current air pollution control devices have little effect on capture Hg⁰ due to its high volatility, low water solubility, and low reactivity.

In recent decades, several technologies have been developed to remove Hg⁰ from flue gas. Among these technologies, activated carbon injection has been regarded as a promising approach for Hg⁰ removal, which obtained more than 90% mercury removal efficiency. However, high maintenance, operation cost and low reproducibility still hinder its industrial application in the developing countries. Therefore, it is necessary to develop the cost-effective alternative materials for elemental mercury removal from coal-fired flue gas.

Sulfur-impregnated carbons and sulfide minerals show a great potential for elemental mercury adsorption. Korpel et al. reported that the sulfur-impregnated activated carbons exhibited the enhanced Hg⁰ removal efficiency due to the easy and stable formation of mercuric sulfide on the carbon surface. Compared with traditional active carbon and chars, the sulfur-impregnated activated carbon was an effective sorbent. The reaction temperature and oxygen content had great influence on mercury removal performance, while SO₂ and NO had no impact on adsorption processing.

Li et al. reported that the Nano-ZnS showed an excellent Hg⁰ adsorption capacity due to the abundance of surface sulfur sites. The activated carbon sorbents modified by metal sulfide were prepared, and their Hg⁰ adsorption capacity increased several times than the activated carbon, with the predominance of chemisorption. The mercury removal performance always depended on the preparation parameters, such as the method of sulfur impregnation, impregnation temperature, and impregnation time. The distribution of elemental sulfur was considered to be the key to develop an effective sorbent. However, the traditional preparation method of the sulfur-impregnated sorbents was complicated and time-consuming, resulting in the obvious increase of Hg removal cost.

More recently, many researchers have committed to investigate on removing mercury from coal-fired flue gas using industrial wastes or byproducts. Li et al. reported that the pyrolyzed biochars from an industrial medicinal residue modified by microwave activation and NH₄Cl impregnation...
captured the elemental mercury efficiently in flue gas. It was considered to be a promising alternative to the commercial activated carbon sorbent for capture of elemental mercury from coal-fired flue gas. Xiao et al.\textsuperscript{16} found that petroleum coke, a waste byproduct of petroleum refining, exhibited the excellent mercury removal performance after being modified by chemical–mechanical bromination. In addition, high sulfur and low cost made the petroleum coke a suitable raw material for mercury capture from coal-fired flue gas. More and more industrial wastes or byproducts, containing elemental sulfur or sulfur compounds, have drawn a substantial amount of attention and are used to remove the mercury from coal-fired flue gas.\textsuperscript{17,18}

At present, land fill is the main processing method to dispose the waste iron oxide desulfurizer. However, there are lots of problems for the method, such as high treatment cost and serious waste of sulfur resource. The work employed a series of used sulfur-containing sorbents for capturing elemental mercury from simulated coal-fired flue gas at low temperatures. The physicochemical properties of samples were characterized by thermogravimetric analysis (TGA), the Brunauer–Emmett–Teller (BET) method, scanning electron microscopy with energy-dispersive spectroscopy (SEM-EDS), X-ray diffraction (XRD), and X-ray photoelectron spectroscopy (XPS). Besides, the effects of temperature and atmosphere on Hg\textsubscript{0} removal efficiency were investigated. The possible mechanism and reaction processing for Hg\textsubscript{0} removal were proposed based on the experimental results and characterization analysis.

2. EXPERIMENTAL SECTION

2.1. Sample Preparation. The Fe/SC catalysts were prepared by the hydrothermal impregnation method. The Fe(NO\textsubscript{3})\textsubscript{3}·9H\textsubscript{2}O was used as a Fe\textsubscript{2}O\textsubscript{3} precursor with semicoke as a supporting material. Table 1 shows the proximate analysis and ultimate analysis of semicoke. The 40–60 mesh SC and the 7wt % Fe(NO\textsubscript{3})\textsubscript{3} solution were placed in an autoclave with hydrothermal impregnation at 200 °C for 5 h. After cooling to room temperature, the samples were filtered and dried at 110 °C for 12 h. Finally, the samples were calcined at 500 °C for 4 h.

The H\textsubscript{2}S selective catalytic oxidation experiments were evaluated on a fixed-bed reactor in a set period at different reaction temperatures (120, 150, 180, and 210 °C). The concentration of H\textsubscript{2}S was 2000 ppm with 0.5 L/min gas flow rate, and the ratio of H\textsubscript{2}S to O\textsubscript{2} in volume content was 2:1. Then, the sorbents were obtained to capture the elemental mercury from simulated coal-fired flue gas. The fresh and used sorbents were denoted fresh-Fe/SC, used-Fe/SC\textsubscript{120}, used-Fe/SC\textsubscript{150}, used-Fe/SC\textsubscript{180}, and used-Fe/SC\textsubscript{210}.

2.2. Sample Characterization. The total sulfur content in samples was tested by 5E-IRS3600 (CKIC, China). The phase structure of samples was determined by an X-ray diffractometer (Bruker, Germany) using Cu k\textsubscript{α} radiation in the range of 10–80° (2\textdegree) with a step size of 0.02°. The specific surface area, pore volume, and average pore size were measured by the physical adsorption/desorption of N\textsubscript{2} at 77 K. The surface atomic states of the sorbents were analyzed by a K\textsubscript{α} X-ray photoelectron spectrometer (Kratos, U.K.) with an Al K\textsubscript{α} X-ray source at}

| Sample | Proximate Analysis (wt %) | Ultimate Analysis (wt %) |
|--------|--------------------------|--------------------------|
|        | Md | Ad | Vad | FCdaf | Cdaf | Hdaf | Odaf* | Ndaf | Sdaf |
| SC     | 6.41 | 7.62 | 30.46 | 69.54 | 79.37 | 3.86 | 15.54 | 0.95 | 0.28 |

Note: ad, air dried basis; d, dry basis; daf, dry and ash free basis. * by difference.
room temperature. The binding energies were calibrated by setting the C 1s peak at 284.8 eV.

2.3. Hg\(^0\) Adsorption and Desorption Experiments. The Hg\(^0\) removal tests were carried out on a fixed-bed reactor device under atmospheric pressure. Figure 1 shows the schematic diagram of the experimental system. The test system contained five parts: a simulated flue gas system, a Hg\(^0\) vapor generator, a fixed-bed reactor, a temperature-controlled tubular furnace, and a continuous online Hg\(^0\) analyzer (Thermo Scientific CEMS). The fixed-bed reactor (inner diameter of 20 mm, length of 50 mm, Quartz glass) was loaded with 500 mg of sorbent samples and placed vertically in a tubular furnace. All of the pipe connecting joints and containers were made of Teflon and covered with heating tapes, so Hg\(^0\) could not be adsorbed on the solid surfaces at a constant temperature of 100 °C. The total flow rate of simulated flue gas was 1.0 L/min, corresponding to a gas hourly space velocity of 15 000 h\(^{-1}\). The initial gaseous Hg\(^0\) concentration in the flue gas was about 60.0 μg/m\(^3\), which was provided by a Dynacal Hg\(^0\) permeation device (VICI Metronics). To guarantee the constant permeating concentration, the Hg\(^0\) permeation was placed in a U-shape glass tube, immersed in a 45 °C water bath.

A series of tests were designed to explore the suitable used sorbent and the impact of different operational parameters on Hg\(^0\) removal. The designed experimental conditions are summarized in Table 2. The running time of each experiment was 90 min. The adsorption test was repeated 3 times with their average values reported. The Hg\(^0\) concentration in the inlet and outlet of the reactor were monitored by CEMS 80i, denoted Hg\(_{in}^0\) and Hg\(_{out}^0\), respectively. The exhaust gas from the mercury analyzer was immediately introduced into the activated carbon trap before discharging into the atmosphere. The Hg\(^0\) removal efficiency, \(E_t\), was defined as

\[
E_t = \frac{Hg_{in}^0 - Hg_{out}^0}{Hg_{in}^0} \times 100\%
\]

Here, Hg\(_{in}^0\) and Hg\(_{out}^0\) are the concentrations in the inlet and outlet of the reactor, respectively.

3. RESULTS AND DISCUSSION

3.1. Characterizations of Samples. TG technology was employed to identify the sulfur forms in the used Fe/SC sorbents. Figure 2a,b shows the TG curves for fresh and used sorbents and for the reaction product sulfur, respectively. The initial mass loss below 150 °C was generally ascribed to the loss of physically and chemically adsorbed water. With the increase of temperature, the weight loss of samples was obviously observed at 200–310 °C. The variation curves of used Fe/SC in Figure 2a were significantly different from those of the fresh one because of elemental sulfur and sulﬁdes in them. The weight loss of used samples was coincident with the mass loss change of the reaction product sulfur exhibited in Figure 2b. The results showed that the weight loss of used Fe/SC should be mainly attributed to the release of elemental sulfur.

The total sulfur contents of the samples fresh-Fe/SC, used-Fe/SC120, used-Fe/SC150, used-Fe/SC180, and used-Fe/SC210 were 0.270, 6.289, 5.715, 5.526, and 5.425%, respectively, which are summarized in Table 3. The contents of sulfur in the used samples were slightly reduced with the
increase of reaction temperature. When the reaction temperature was low (e.g., less than 120 °C), a generous amount of reaction product elemental sulfur was left in the samples. As the reaction temperature increased, it was more conducive to the reaction of H₂S with Fe₂O₃ to form FeS, resulting in decrease of sulfur selectivity. Simultaneously, sulfur product released into the gas in the form of gaseous sulfur more easily.

The BET surface areas, pore volumes, and average pore diameters of different samples are presented in Table 3. The specific surface area of fresh-Fe/SC was 321.281 m²/g. The specific surface areas of used-Fe/SC120, used-Fe/SC150, used-Fe/SC180, and used-Fe/SC210 decreased to 153.645, 101.217, 47.096, and 32.339 m²/g, respectively.

Figure 3 illustrates that micropores dominated in the fresh-Fe/SC derived from lignite semicoke and the pore sizes were mainly distributed in the regime of 0.4 ≤ d ≤ 1.5 nm. With the increase of catalytic oxidation temperature of H₂S, the specific surface area of used sorbent decreased rapidly. In addition, the content of micro-mesoporous structure and the N₂ adsorption capacity in the used sorbents decreased obviously. The reasons accounting for this phenomenon might be due to the elemental sulfur deposition, channel plugging, and the growth of FeS particle size after the reaction.

To reveal the surface structures of used sorbents at different reaction temperatures, the texture and morphology of used-Fe/SC120 and used-Fe/SC210 were investigated by SEM, which are illustrated in Figure 4a,b. Figure 4 shows that the surface morphology of used-Fe/SC120 was different from that of used-Fe/SC210. The pore structures of the used-Fe/SC120 did not block the pores and channels as serious as that of used-Fe/SC210, on the surface of which the FeS particle agglomeration occurred. Therefore, the specific surface area of used-Fe/SC120 was much larger than that of used-Fe/SC210.

Figure 4c,d shows that the surface of used-Fe/SC120 was covered with the generated sulfur, which appeared irregular with a needlelike morphology. The sulfur was attached to the sorbent surface, which provided more S active sites to facilitate Hg⁰ absorption. However, the elemental sulfur was just attached to the surface of the sorbent with poor interface combination ability. Nevertheless, the EDS analysis in Figure 4e indicated that partial Fe existed in the form of FeS. However, the diffraction peaks corresponding to FeS and elemental sulfur did not stand out. This result was attributed to the fact that FeS and elemental sulfur were in an amorphous or poorly crystalline state.

3.2. Hg⁰ Adsorption Performance. 3.2.1. Comparisons of Hg⁰ Removal Performance of Fresh and Used Fe/SC. Figure 6 shows the Hg⁰ removal efficiency of different used sorbents at 120 °C. It could be observed that the Hg⁰ removal performance of sorbents decreased in different degrees. The Hg⁰ removal performances of fresh-Fe/SC, used-Fe/SC180 and used-Fe/SC210 were similar. The instantaneous Hg⁰ removal efficiency was about 80%, and then decreased gradually to about 63.3%. The removal performance of used-Fe/SC120 was slightly higher than that of used-Fe/SC150 and significantly much higher than that of the other three. The mercury removal efficiency of used-Fe/SC120 decreased slowly from 93.2 to 79.6% within 90 min. This might be attributed to the fact that used-Fe/SC120 had quite rich pore structure and more S active sites on the surface of the sorbent.
Due to the higher specific surface and the developed pore structure, the fresh-Fe/SC exhibited good mercury removal performance. The \( \text{Hg}^0 \) removal over used-Fe/SC occurred through chemical adsorption, while the \( \text{Hg}^0 \) removal over fresh-Fe/SC occurred through physical adsorption. Thus, it could be concluded that the higher surface area and more S active sites were both essential for a better \( \text{Hg}^0 \) removal performance.

3.2.2. Effect of Adsorption Temperature. Used-Fe/SC120 was chosen to investigate the impact of the reaction temperature on mercury removal performance. Figure 7a displays the \( \text{Hg}^0 \) removal efficiency of used-Fe/SC120 at the temperature range from 60 to 150 °C. Figure 7a shows that temperature had great influence on the \( \text{Hg}^0 \) removal efficiency and that \( \text{Hg}^0 \) adsorption significantly weakened with the increase of temperature. The mercury removal efficiency was stable within 90 min at 60 and 90 °C, with the average efficiencies of 92.4 and 89.5%, respectively. However, the \( \text{Hg}^0 \) adsorption performance of used-Fe/SC120 decreased with the temperature increasing to 120 °C. At 150 °C, the \( \text{Hg}^0 \) adsorption efficiency dropped sharply from the initial 88.5 to 44.4% within 90 min. It might be attributed to the fact that a lower temperature was more suitable for the reaction between S active sites and \( \text{Hg}^0 \). Nevertheless, the decrease of \( \text{Hg}^0 \) removal efficiency at 150 °C was due to the desorption of \( \text{Hg}^0 \) adsorbed on the surface of sorbent.

The inhibition of \( \text{Hg}^0 \) adsorption at 150 °C might be interpreted by another possible reason: The release of elemental sulfur from the sorbent decreased the surface sulfur coverage. The effect of adsorption temperature on surface sulfur content was displayed in Figure 7b. Some surface sulfur released from sorbents after experiment at 60 and 90 °C. The release of surface sulfur occurred and become more serious with the adsorption temperature increasing to 150 °C. The reason was that the high temperature promoted the reaction of S and \( \text{O}_2 \). As a result, the decrease of surface sulfur content reduced the S active sites.
3.2.3. Effect of Flue Gas Components. Figure 8 shows that in the absence of O2 the Hg0 removal average efficiency was about 91.27%, while the average efficiency decreased to about 86.9% with 4% O2 being introduced into the reaction system. With the O2 concentration increasing to 8%, the average efficiency decreased to about 82.8%. This suggested that O2 had a slight adverse effect on Hg0 adsorption over used-Fe/SC120 at 90°C. A small amount of unstable surface sulfur reacted with O2 to form SO2, resulting in the decrease of the amount of active sites. SO2 played an inhibitive role in Hg0 adsorption as well. The Hg0 removal average efficiency decreased from 86.9 to 83.1 and 80.8% with the addition of 500 and 1000 ppm SO2 into a 4%O2 + N2 + 10%CO2 atmosphere, respectively. This could be attributed to the competitive adsorption between SO2 and Hg0 on the surface of sorbents. When 300 ppm NO was added into the 4%O2 + N2 + 10%CO2 atmosphere, the average efficiency decreased slightly from 86.9 to 85.9%. Then, it increased NO concentration to
600 ppm, with the Hg\textsuperscript{0} removal efficiency improved slightly. The results suggested that there was a possible reaction route between NO\textsubscript{2} and Hg\textsuperscript{0} to produce HgO and NO.

The stability experiment of the used-Fe/SC120 sorbent for Hg\textsuperscript{0} removal was carried out, and Figure 9 shows the results. The Hg\textsuperscript{0} removal efficiency decreased from 93.3 to 79.5\% within 360 min, implying that the used-Fe/SC120 sorbent was an effective sorbent for Hg\textsuperscript{0} capture.

3.2.4. Mechanism of Hg\textsuperscript{0} Removal. XPS analysis was employed to explore the chemical state and the relative portion of Fe, S, and Hg on the surface of sorbents before and after Hg\textsuperscript{0} removal, and the results are shown in Figure 10.

It can be seen from Figure 10a that the Fe 2p spectra for fresh and used sorbents showed three peaks at 710.5, 711.7, and 713.0 eV, corresponding to Fe\textsuperscript{2+} species bonded with S\textsuperscript{2−}, Fe\textsuperscript{3+} species bonded with O\textsuperscript{2−}, and Fe\textsuperscript{3+} species bonded with SO\textsubscript{4}\textsuperscript{2−}, respectively.\textsuperscript{19–21} The Fe 2p peaks for the surface of the fresh and used sorbents were basically the same, suggesting that Fe had not participated in the Hg\textsuperscript{0} adsorption process.

The S 2p spectra of the fresh and used sorbents are shown in Figure 10b. The peaks at 164.0, 164.9, and 168.9 eV were assigned to polysulfur and S species in S\textsuperscript{2−} and SO\textsubscript{4}\textsuperscript{2−}, respectively.\textsuperscript{22,23} The sulfate was not supposed to participate in the process of mercury removal, taking the peak area of sulfate at 168.9 eV as a reference point. The peak area of polysulfur in the sorbent decreased significantly, with the ratio of polysulfur/sulfate decreased from 1.696 to 0.611. This might be ascribed to the fact that surface sulfur reacted with Hg\textsuperscript{0} to form HgS.

Figure 10c shows the XPS spectrum of Hg 4f for used-Fe/SC120 after Hg\textsuperscript{0} removal tests. The peak at 100.9 eV was ascribed to HgS,\textsuperscript{24,25} which formed from the reaction of elemental mercury and sulfur. This further confirmed that chemical adsorption occurred on the surface S active sites of the sorbent.

The method of temperature-programmed decomposition (TPD) was widely used to identify the occurrence modes of Hg in coals.\textsuperscript{26–28} Hg-TPD was used to identify the mercury species in the used sorbent, and Figure 11 shows the desorption profiles of Hg. There was a well-resolved peak at about 200 °C, which could be ascribed to the release of β-HgS.\textsuperscript{29,30}

Therefore, the possible mechanism for Hg\textsuperscript{0} removal over the used-Fe/SC120 at low temperatures may be described as

\[
\text{Hg}^0(\text{g}) \rightarrow \text{Hg}^0(\text{ad})
\]
The presence of \( \text{O}_2 \) obviously was not conducive to \( \text{Hg}^0 \) removal. \( \text{SO}_2 \) played an inhibitive role in \( \text{Hg}^0 \) removal experiment.

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