Biomass Carbon Magnetic Adsorbent Constructed by One-Step Activation Method for the Removal of Hg⁰ in Flue Gas

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ABSTRACT: Elemental mercury (Hg⁰) emission from industrial boilers equipped in factories such as coal-fired power plants poses serious hazards to the environment and human health. Herein, an iron-modified biomass carbon (Fe/BC) magnetic adsorbent was prepared by a one-step method using pepper straw waste as raw material and potassium oxalate and ferric nitrate as activator and catalyst precursor, respectively. A fixed-bed reactor was used to evaluate the Hg⁰ removal performance of the Fe/BC adsorbent. The synthesized adsorbent showed a wide temperature window for Hg⁰ removal. In a N₂ + O₂ atmosphere, the removal efficiency toward Hg⁰ was 97.6% at 150 °C. Further, O₂ or SO₂ could promote the removal of Hg⁰, while NO could inhibit the conversion of Hg⁰ over the Fe/BC adsorbent. The consequence of XPS and Hg-TPD showed that lattice oxygen in Fe₂O₃ and chemisorbed oxygen were the main active sites for Hg⁰ removal, and HgO was the main mercury species on used Fe/BC. Moreover, Fe/BC adsorbent showed a good regeneration and magnetization performance, which was conducive to the cost reduction of actual industrial application. This study provides a facile approach for efficient removal of Hg⁰ using biomass-derived carbon material.

1. INTRODUCTION

Mercury (Hg), an important lethal pollutant, is a trace heavy metal element and displays toxicity, for instance, persistence, mobility, and bioaccumulation in both food chains and ecosystems.¹ ² The anthropogenic Hg emissions basically originate from coal combustion. The proportion of it in total emissions is around 30%.³ There are three forms of mercury in flue gas: mercury oxide (Hg²⁺), particle bound mercury (Hgₚ), and elemental mercury (Hg⁰).⁴⁻⁶ It is well-known that Hg⁰ and Hg²⁺ can be easily controlled by dust removal devices and wet flue gas desulfurization (WFGD), respectively.⁷ ⁸ Nevertheless, Hg⁰ is difficult to remove because it is insoluble and volatile at normal temperature and pressure.⁹ ¹⁰ Consequently, development of systems that ensure the removal of Hg⁰ is imperative.

In recent years, for the removal efficiency of Hg⁰, substantial related technologies from adsorption¹¹ to catalytic oxidation¹² have been applied. Adsorption capture is considered as one of the most promising ones in all technologies. For the removal of Hg⁰ in coal combustion flue gas, the adsorbents have been accessed by many investigators, for example, activated carbons (AC),¹¹ ¹² mineral materials,¹³ zeolites,¹⁴ calcium sorbents,¹⁵ petroleum coke,¹⁶ and fly ash.¹⁷ Among these, AC has been regarded as one of the most effective materials for the removal of Hg⁰. However, the high operating cost of conventional AC limits its large-scale applications in power plants. Therefore, discussions regarding the development of low-cost adsorbents have become a dominant research area in recent years.

As a renewable resource, biomass has been widely applied in the preparation of carbon-based adsorbents for catalysis and adsorption reactions because of its abundance, sustainability, economic benefits, and environmentally benign nature.¹⁹ According to statistics, China’s pepper planting area ranks second among vegetables. Because of its high carbon content and low ash content, this area has the potential to prepare biomass carbon.²⁰ Pepper straw also contains different phenolic, carboxyl, ether, and amine groups, which may adsorb toxic elements in the environment.²¹ Therefore, the use of pepper straw for biomass carbon production is a feasible approach in terms of industrial waste management and renewable material development for Hg⁰ elimination from flue gas.

Raw biomass carbon shows a low Hg⁰ adsorption capacity because of its poor surface active sites. The surface active sites of biomass carbon can be improved by chemical means so as to
effectively improve the adsorption performance of Hg. Chemical activation can introduce active groups on the biomass carbon via acid, alkali, metal, sulfur, and halogen modifications, which promotes the removal of Hg. Although the biomass adsorbents can be used as viable substitutes for AC, the powdered biomass carbon injected into flue gas will be captured by dust control devices together with fly ash, which is not conducive to the reuse of fly ash. Recently, to more efficiently recycle the used adsorbent from fly ash, some cheap magnetic mercury sorbents have been developed. For example, Yang et al. combined magnet cobalt iron impregnated porous carbon, and the active components of Hg capture were chemisorbed oxygen and lattice oxygen derived from CO3O4, Fe3O4, and Fe2O3. Xu et al. claimed that the preparation of a magnet organic carbon adsorbent by one-step pyrolysis of organic matter containing FeCl3 showed that Fe3O4 and or Cl− could accurately improve the removal rate of Hg. Shan et al. compounded a magnetic Mn–Fe biomass-based carbon adsorbent for capturing Hg from flue gas, and chemisorbed oxygen, lattice oxygen and active species on adsorbent surface were profitable for Hg removal. This research further inspired us to explore new methods of preparing magnetic biomass-based adsorbents through a simple preparation process that offers low cost and good regeneration performance to meet practical industrial applications. Ferrous nitrate Fe(NO3)3 is a safe and cheap chemical reagent that is commonly used as an oxidant in organic synthesis and a precursor of Fe3O4/Fe(NO3)3 as the activator and catalyst precursor, respectively.

2.2. Preparation of Iron-Modified Biomass Carbon. Fe/BC adsorbents were prepared via a chemical activation method. K2C2O4 was selected as an activator. K2C2O4 decomposed into K2CO3 at low temperature, and the generated K2CO3 further corroded carbon. Further, as a form-poring agent, the released CO is devoted to development of the porous structure. During the impregnation process of activator K2C2O4, Fe(NO3)3 was simultaneously added to prepare Fe/BC. This one-step activation and modification could reduce the times of calcination. The specific steps are as follows: First, pepper straw (3 g) was thoroughly mixed with an appropriate amount of K2C2O4 solution and Fe(NO3)3 solution, and then the mixed material solution was allowed to sit at room temperature for 10 h. Second, based on a certain temperature procedure and N2 atmosphere, the mixture was treated in a tube furnace; the mixture was heated to 30 °C directly and maintained at that temperature for 2 h before being cooled. The treated sample was ultimately washed with deionized until the pH was close to 7. The sample was dried at 80 °C for 12 h. The prepared Fe/BC adsorbent was named A0.075-Fe0.4-T850, where x and y represent the concentrations of K2C2O4 and Fe(NO3)3 solutions, respectively; T represents the activation temperature; and z represents the specific temperature. For example, A0.075-Fe0.4-T850 adsorbent indicates that pepper straw was activated at 850 °C and K2C2O4/Fe(NO3)3 = 0.075:0.4 (concentration ratio).

2.3. Elemental Mercury Removal Experiments of Iron-Modified Biomass Carbon Adsorbents. The tests for Hg removal of Fe/BC adsorbents can be accessed in a fixed-bed system at laboratory scale. The evaluation apparatus reported in our previous study was used herein. In the test steps, the total simulated flue gas flow was controlled at 1000 mL min−1. The flue gas flow was 4 vol % O2, 0.02 vol % SO2 (when used), and 0.04 vol % NO (when used), 40 ± 2 μg m−3 Hg vapor, and N2. Through the precision of the mass flow controller, the flow rate of each gas was easily controlled. The Fe/BC adsorbent (0.16 g, 0.8 mL) was injected into a quartz reactor with a diameter of 8.0 mm. Then the simulated flue gas was brought into the reactor at an estimated temperature.

At the inside and outside of this reactor, the Hg concentrations were used in the Hg analyzer. Because of the removal performance of Hg0, the adsorbents’ activity was measured (η). The performance could be calculated following eq 1:

\[ \eta = \frac{C_{in} - C_{out}}{C_{in}} \times 100 \]  

where \( C_{in} \) and \( C_{out} \) are the inside and outside concentrations (μg m−3) of Hg0.

2.4. Characterization. By using the Micromeritics ASAP-2460 analyzer to pass the N2 adsorption–desorption test at −196 °C, the pore characteristics of the Fe/BC adsorbent...
Fe(NO$_3$)$_3$) exhibited a rather low Hg$^0$ removal performance of about 0.4% in 2 h. The removal efficiency of adsorbent for Hg$^0$ was improved at different levels after chemical activation. The removal efficiency of sample A$_{0.075}$-Fe$_{0.4}$-T800 was lower than other samples. As the activation temperature increased from 750 to 800 and 850 °C, the removal efficiency of Hg$^0$ dropped from 97.6% to 44.2%. This result indicated that the sample carbonized at 850 °C exhibited the highest removal efficiency of Hg$^0$. Thus, A$_{0.075}$-Fe$_{0.4}$-T850 adsorbents were used for subsequent experiments.

3. RESULTS AND DISCUSSION

3.1. Elemental Mercury Capture. 3.1.1. Effect of Preparation Conditions on Elemental Mercury Removal.

The removal performance of Hg$^0$ of Fe/BC magnetic adsorbents, which were prepared at different doses of activator K$_2$C$_2$O$_4$ and catalyst precursor Fe(NO$_3$)$_3$, was assessed in a N$_2$−O$_2$−Hg atmosphere at 150 °C. Figure 1a shows the results, and the sample A$_0$-Fe$_0$-T800 (without K$_2$C$_2$O$_4$ and Fe(NO$_3$)$_3$) exhibited a rather low Hg$^0$ removal performance of about 0.4% in 2 h. The removal efficiency of adsorbent for Hg$^0$ improved at different levels after chemical activation. The Hg$^0$ removal efficiency of sample A$_{0.075}$-Fe$_{0.4}$-T800 decreased from 51.3% to 10.1% within 2 h. These results showed that the removal activity of the Hg$^0$ of the adsorbent prepared with the addition of K$_2$C$_2$O$_4$ activator was better than that of the adsorbent without the addition of any activator in 2 h. The removal performance of Hg$^0$ of the sample A$_0$-Fe$_{0.4}$-T800 decreased from 55.0% to 40.1%, indicating that the removal efficiency of Hg$^0$ of the adsorbent prepared using only Fe(NO$_3$)$_3$ was better than that using only K$_2$C$_2$O$_4$ as activator. The Hg$^0$ removal efficiency of sample A$_{0.075}$-Fe$_{0.4}$-T800 was maintained at above 75.9% within 2 h. The results showed that the adsorbent prepared by adding K$_2$C$_2$O$_4$ and Fe(NO$_3$)$_3$ exhibited the best Hg$^0$ removal activity; the reason may be that K$_2$C$_2$O$_4$ contributes to the pore development, and Fe(NO$_3$)$_3$ was decomposed into Fe$_3$O$_4$ during activation process, which had good mercury removal ability.

Figure 1b shows the effect of activation temperature (700, 750, 800, 850, and 900 °C) on the removal efficiency of Hg$^0$ of Fe/BC. It can be seen that the removal performance of Hg$^0$ of sample A$_{0.075}$-Fe$_{0.4}$-T700 was lower than other samples. As the activation temperature increased from 750 to 800 and 850 °C, the removal efficiency of Hg$^0$ increased from 44.1% to 75.9% and 97.6%, respectively. The experimental results showed that the high activation temperature had an apparent impact on the removal of Hg$^0$. However, when the activation temperature gradually increased from 850 to 900 °C, the removal efficiency of Hg$^0$ dropped from 97.6% to 44.2%. This result indicated that the sample carbonized at 850 °C exhibited the highest removal efficiency of Hg$^0$. Thus, A$_{0.075}$-Fe$_{0.4}$-T850 adsorbents were used for subsequent experiments.

3.1.2. Effect of Adsorption Temperature on Elemental Mercury Removal. Hg$^0$ removal experiments were conducted at five different temperatures to analyze the effect of adsorption temperature on Hg$^0$ removal efficiency, i.e., 60, 90, 120, 150, and 180 °C. Figure 1c shows the results. It can be seen that the removal efficiency of Hg$^0$ of Fe/BC adsorbent increased with an increase in temperature from 60 to 150 °C. At 150 °C, the removal efficiency of Hg$^0$ was the highest (97.6%). Xie et al. claimed that a higher temperature was beneficial for accelerating the chemical reaction rate between Hg$^0$ and the active sites on the adsorbent. Nevertheless, the removal performance of Hg$^0$ of A$_{0.075}$-Fe$_{0.4}$-T850 adsorbent was lower than at 180 °C than at 150 °C. The performance decreased from 100% to 60.5% within 2 h. Yang et al. proved that the...
physical adsorption of Hg\textsuperscript{0} could not get assistance from excessive temperature and could even cause a release of the captured Hg\textsuperscript{0} from the surface of adsorbents, thereby weakening Hg\textsuperscript{0} removal. The above-mentioned experimental results indicated that the A\textsubscript{0.075-Fe0.4-T850} adsorbent exhibited a wide temperature window of 60\textdegree C to 150\textdegree C, and 150\textdegree C was considered as the optimal reaction temperature.

3.1.3. Effect of Simulated Gas Components on Elemental Mercury Removal. On this basis, the Hg\textsuperscript{0} capture process is usually carried out in multicomponent flue gas. Thus, it is necessary to analyze the impact of flue gas components on Hg\textsuperscript{0} adsorption. The impact of two main acid gas components (SO\textsubscript{2} and NO) on the removal of Hg\textsuperscript{0} by A\textsubscript{0.075-Fe0.4-T850} adsorbent was analyzed at 150\textdegree C. The impacts of SO\textsubscript{2} and NO were analyzed when the oxygen volume fraction was 4%. Figure 1d shows the results. The Hg\textsuperscript{0} removal efficiency of A\textsubscript{0.075-Fe0.4-T850} adsorbent in N\textsubscript{2} atmosphere decreased from 86.9\% to 55.3\% within 2 h. Under N\textsubscript{2} + O\textsubscript{2} atmosphere, the removal efficiency of Hg\textsuperscript{0} reached 97.6\% within 2 h. The removal efficiency of Hg\textsuperscript{0} in N\textsubscript{2} + O\textsubscript{2} + NO atmosphere was lower than that in N\textsubscript{2} + O\textsubscript{2} atmosphere, which indicated that NO could inhibit the Hg\textsuperscript{0} removal. It can be seen from Figure 1e that the Hg\textsuperscript{0} removal efficiency gradually decreased with the addition of NO from 200 to 800 ppm. In order to explore the inhibition mechanism of NO on the Hg\textsuperscript{0} removal performance of adsorbents, NO was intermittently injected into the N\textsubscript{2} + O\textsubscript{2} atmosphere during the evaluation of Hg\textsuperscript{0} removal efficiency of the adsorbent at 150 \textdegree C, and the change trend of the adsorbent Hg\textsuperscript{0} removal was recorded. It can be seen from Figure S1 that the mercury removal efficiency of the A\textsubscript{0.075-Fe0.4} sample significantly decreased. When NO was cut off, the Hg\textsuperscript{0} removal efficiency of the A\textsubscript{0.075-Fe0.4} adsorbent increased. This could be attributed to competitive adsorption occurring between Hg\textsuperscript{0} and NO on the active site of the adsorbent.

The impact of SO\textsubscript{2} on Hg\textsuperscript{0} removal was difficult to explain because it may depend on the surface characteristics of adsorbent or flue gas components.\textsuperscript{48} Figure 1f shows that the introduction of SO\textsubscript{2} promoted the removal of Hg\textsuperscript{0}. The removal efficiency of Hg\textsuperscript{0} was 100\% within 2 h. When the SO\textsubscript{2} concentration was 200, 400, 600, and 800 ppm, respectively, the Hg\textsuperscript{0} removal efficiency of the Fe/BC adsorbent remained 100\% within 2 h. The existence of SO\textsubscript{2} improved the removal efficiency of Hg\textsuperscript{0} of Fe/BC adsorbent. It can be seen from Figure 7 that there was a mercury release peak at 203 \textdegree C after the introduction of SO\textsubscript{2}, which was attributed to \(\beta\)-HgS.\textsuperscript{49} Figure 6d shows the S 2p XPS spectra of fresh and used A\textsubscript{0.075-Fe0.4} samples. The peak at 169.4/169.3 eV was assigned to SO\textsubscript{4}\textsuperscript{2-}, and the peak at 164.4 eV was assigned to active sulfur.\textsuperscript{50,51} It can be seen that the signal of sulfur was very weak in the fresh sample, and the fresh sample did not show the peak at 164.4 eV. However, the used adsorbent in SO\textsubscript{2} atmosphere presented active sulfur at 164.4 eV, which was beneficial for mercury removal.\textsuperscript{50,51} Therefore, the effects of SO\textsubscript{2} on the removal of Hg\textsuperscript{0} over Fe/BC adsorbents can be concluded: SO\textsubscript{2} was adsorbed on the surface of Fe/BC adsorbents, and could dissociate active sulfur sites and oxygen sites on the surface of Fe\textsubscript{2}O\textsubscript{3}.\textsuperscript{52} Finally, the dissociated active sulfur on the surface of Fe/BC adsorbents could react with Hg to form \(\beta\)-HgS.\textsuperscript{49}

3.1.4. Comparison of the Fe/BC Adsorbents with Other ACs/Modified ACs. The characteristics of some magnetic

![Figure 2.](https://doi.org/10.1021/acsomega.1c05857)
Hg\textsubscript{0} removal of Fe/BC adsorbent. The removal of Hg\textsubscript{0} from 750 to 900 °C showed greater capability of Hg\textsubscript{0} capture than the others. Addition of only K\textsubscript{2}C\textsubscript{2}O\textsubscript{4} mainly resulted in the generation of Fe\textsuperscript{2+}O\textsubscript{3}, and the Fe\textsuperscript{2+}O\textsubscript{3} was reduced by CO and gradually was transformed into Fe\textsuperscript{0}.42 The element Fe could be explained as follows: K\textsubscript{2}C\textsubscript{2}O\textsubscript{4} decomposed into K\textsubscript{2}CO\textsubscript{3} at 600 °C, and the generated K\textsubscript{2}CO\textsubscript{3} further corroded carbon and then released CO. In the process of carbonization, Fe(NO\textsubscript{3})\textsubscript{3} was resolved to Fe\textsubscript{2}O\textsubscript{3}, and the Fe\textsubscript{2}O\textsubscript{3} was reduced by CO and gradually was transformed into Fe\textsuperscript{0}.42 The iron-modified porous carbon was prepared by the one-step method and the adsorbents prepared have high Hg\textsubscript{0} adsorption performance and magnetic performance.

3.2. Characteristics of Iron-Modified Biomass Carbon Adsorbent. 3.2.1. Analysis of Pore Property. In order to analyze the texture characteristics of Fe/BC adsorbent, the N\textsubscript{2} adsorption desorption isotherm of the adsorbent was measured. Figure 2 shows the results. Table 2 lists the BET surface area (S\textsubscript{BET}), average pore diameter (D\textsubscript{p}), and pore volume (V\textsubscript{p}). Parts a and c of Figure 2 demonstrated that N\textsubscript{2} gas adsorption existed in the Hg\textsubscript{0} removal process. The regenerated Fe/BC adsorbent remained as a good superparamagnetic structure after the four cycles and its saturation magnetization remained at 16.7 emu g\textsuperscript{-1}, which promoted the recovery of the adsorbent after use. The preparation methods reported in the literature were relatively complicated. In this work, the magnetic Fe-modified biomass-based activated carbon, Fe/BC adsorbent showed good Hg\textsubscript{0} removal performance of Hg\textsubscript{0} (about 0.4%).

Table 2. Pore Structure Characteristics of Fe/BC Adsorbents

| sample name | surface area (m\textsuperscript{2} g\textsuperscript{-1}) | V\textsubscript{p} (cm\textsuperscript{3} g\textsuperscript{-1}) | D\textsubscript{p} (nm) |
|-------------|-----------------|-----------------|-----------------|
| A\textsubscript{0}-Fe\textsubscript{0}-T800 | 3.95            | 0.02            | 16.71           |
| A\textsubscript{0.075}-Fe\textsubscript{0.4}-T800 | 913.08          | 0.40            | 1.76            |
| A\textsubscript{0}-Fe\textsubscript{0.4}-T800 | 315.43          | 0.37            | 4.80            |
| A\textsubscript{0.075}-Fe\textsubscript{0.4}-T800 | 543.15          | 0.34            | 2.53            |
| A\textsubscript{0.075}-Fe\textsubscript{0.25}-T800 | 394.35          | 0.22            | 2.19            |
| A\textsubscript{0.075}-Fe\textsubscript{0.75}-T750 | 431.03          | 0.23            | 2.26            |
| A\textsubscript{0.075}-Fe\textsubscript{0.75}-T850 | 428.72          | 0.30            | 2.83            |
| A\textsubscript{0.075}-Fe\textsubscript{0.75}-T900 | 379.09          | 0.34            | 3.62            |

Figure 3 shows that powder XRD patterns of A\textsubscript{0}-Fe\textsubscript{0}-T800 adsorbent and A\textsubscript{0.075}-Fe\textsubscript{0.4}-T850 adsorbents at different activation temperatures were obtained. The diffraction peaks of A\textsubscript{0}-Fe\textsubscript{0}-T800 sorbent at 28.4°, 40.6°, 50.3°, and 66.5° were attributed to KCl. It is relevant to the high ash content of raw pepper straw. Figure 3 showed the presence of obvious characteristic diffraction peaks of five types of Fe/BC samples at 44.7°, and it corresponded to elemental Fe.52 The source of elemental Fe could be explained as follows: K\textsubscript{2}C\textsubscript{2}O\textsubscript{4} decomposed into K\textsubscript{2}CO\textsubscript{3} at 600 °C, and the generated K\textsubscript{2}CO\textsubscript{3} further corroded carbon and then released CO. In the process of carbonization, Fe(NO\textsubscript{3})\textsubscript{3} was resolved to Fe\textsubscript{2}O\textsubscript{3}, and the Fe\textsubscript{2}O\textsubscript{3} was reduced by CO and gradually was transformed into Fe\textsuperscript{0}.42 The diffraction peaks of A\textsubscript{0.075}-Fe\textsubscript{0.4}-T850 adsorbent at 30.5°, 35.9°, 43.4°, 57.5°, and 63.2° were attributed to Fe\textsubscript{2}O\textsubscript{3}. The distinctive peak intensity of Fe decreased little by little as the temperature gradually increased from 750 to 900 °C. A peak temperature of 850 °C was found for graphite carbon. It can also be seen from Figure S2 that the full width at half maxima of G peak of adsorbent decreased and that of D peak increased at 850 °C, compared with other temperatures. That was indicated the graphitization degree of adsorbent increased. This result could explain why the S\textsubscript{BET} of sample A\textsubscript{0.075}-Fe\textsubscript{0.4}-T850 was lower than that of sample A\textsubscript{0.075}-Fe\textsubscript{0.4}-T800,54 which was consistent with the result of XRD.

3.2.2. Analysis of X-ray Diffraction. Figure 3 shows that powder XRD patterns of A\textsubscript{a}-Fe\textsubscript{a}-T800 adsorbent and A\textsubscript{0.075}-Fe\textsubscript{0.4}-T850 adsorbent. The XRD patterns of raw pepper straw and Fe/BC under different activation temperatures.
For further studying the magnetic performance of $A_{0.075}$-$Fe_{0.4}$-T850 sample, VSM was used to obtain and evaluate the magnetization curve of the sample. Figure 4b demonstrated that the Fe/BC adsorbent showed a coercivity in the smallest degree and a magnetization hysteresis which could be ignored, thus indicating that it could be regarded as a super-

Figure 4. Magnetization characteristics: experimental photographs of $A_{0.075}$-$Fe_{0.4}$-T850 sample under an external magnetic field (a); $A_{0.075}$-$Fe_{0.4}$-T850, used $A_{0.075}$-$Fe_{0.4}$-T850, and fourth-regeneration adsorbents (b).

Figure 5. SEM images of (a) $A_{0}$-$Fe_{0}$-T800 and (b) $A_{0.075}$-$Fe_{0.4}$-T800; (c) $A_{0}$-$Fe_{0}$-T850; and (d) $A_{0.075}$-$Fe_{0.4}$-T850 and EDS spectra of (e) $A_{0}$-$Fe_{0}$-T800 and (f) $A_{0.075}$-$Fe_{0.4}$-T800; (g) $A_{0}$-$Fe_{0}$-T850; and (h) $A_{0.075}$-$Fe_{0.4}$-T850.

Figure 6. XPS analysis of fresh and used Fe/BC adsorbents: (a) O 1s, (b) Fe 2p, (c) Hg 4f, and (d) S 2p.
paramagnetic material. The adsorbent Fresh-A$_{0.075}$Fe$_{0.4}$T850 showed good magnetization and saturation magnetization of 25 emu g$^{-1}$. There was no obvious variation of the magnetism between Used-A$_{0.075}$Fe$_{0.4}$T850 sample and Fresh-A$_{0.075}$Fe$_{0.4}$T850 sample. After four regenerations, the magnetism of A$_{0.075}$Fe$_{0.4}$T850 adsorbent was found to weaken. The magnetization property could prevent the sample from being permanently magnetized, which was conducive to the redispersion of sample without the external magnetic field.\textsuperscript{35} This result indicated that when an external magnetic field was introduced, it was possible to recover the used A$_{0.075}$Fe$_{0.4}$T850 adsorbent from fly ash.

3.2.4. Scanning Electron Microscopy Analysis. Figure 5 present the SEM images and EDS spectra of A$_0$-Fe$_0$T800, A$_{0.075}$Fe$_{0.4}$T800, A$_0$-Fe$_0$T850, and A$_{0.075}$Fe$_{0.4}$T850. Compared to A$_0$-Fe$_0$ sample, the surface of the activated sample was loose and porous, which was beneficial for Hg$^0$ removal ability of adsorbents.\textsuperscript{35} It can be seen from the EDS spectra in Figure 5(h) that the samples activated by Fe(NO$_3$)$_3$ mainly contained the elements of C, O, and Fe. In contrast, the composition of the samples without Fe(NO$_3$)$_3$ treatment was C, K, Cl, Si, and O.

3.3. Mechanism Discussion. 3.3.1. Analysis of X-ray Photoelectron Spectroscopy. To clarify the removal mechanism of Hg$^0$ of the Fe/BC adsorbent, the analysis of XPS was used to decide the valence states of the surface elements of the fresh and used A$_{0.075}$Fe$_{0.4}$T850 adsorbent. Figure 6 shows the XPS spectra of O 1s, Fe 2p, Hg 4f, and S 2p. The O 1s XPS spectra of O 1s, Fe 2p, Hg 4f, and S 2p. The O 1s XPS spectrum of fresh and used A$_{0.075}$Fe$_{0.4}$T850 adsorbent is presented in Figure 6a. It shows three peaks for fresh and used adsorbents. Approximately 530.4 eV was accessed to the characteristic peak of lattice oxygen (O\textsubscript{\alpha}). The peak at 531.8/531.7 eV was contributed to chemically adsorbed oxygen (O\textsubscript{\beta}), and the peak at 532.6 eV was contributed to molecular water (O\textsubscript{\gamma}). Yang et al.\textsuperscript{46} suggested that the existence of O\textsubscript{\beta} may be relevant to the charge imbalance, vacancies and chemical bonds produced by metal oxides. Table 3 make a summary after the removal experiment of Hg$^0$. The proportion of O\textsubscript{\beta} increased from 28.6% to 30.5%, while the proportion of O\textsubscript{\gamma} reduced from 46.9% to 36.1%. These data indicated that O\textsubscript{\beta} plays important role for Hg$^0$ removal.

Figure 6b shows the Fe 2p XPS spectra of fresh and used A$_{0.075}$Fe$_{0.4}$T850 samples. The three subpeaks at 710.6, 711.6, and 714.1/713.9 eV (belonging to Fe 2p3/2) were correspond to Fe$^{2+}$, Fe$^{3+}$ (octahedral), and Fe$^{3+}$ (tetrahedral), respectively.\textsuperscript{46} The ratio of Fe$^{2+}$/Fe$^{3+}$ concentration dropped from 6.0% to 4.6% (Table 3) after removal of Hg$^0$. The results showed that Fe$^{3+}$ was reduced to Fe$^{2+}$ during the Hg$^0$ capture process.

Figure 6c shows the results. It was the Hg 4f XPS spectrum of this fresh and used A$_{0.075}$Fe$_{0.4}$T850 sample. The peak centered at 99.6 eV was designated as Hg$^0$, and the peak centered at 101.9 eV was designated HgO.\textsuperscript{55} Thus, it could be inferred that O\textsubscript{\beta} and O\textsubscript{\gamma} were important in the Hg$^0$ removal by the A$_{0.075}$Fe$_{0.4}$T850 adsorbent.

3.3.2. Analysis of Hg-Temperature-Programmed Desorption. To identify the types of Hg adsorbed on the A$_{0.075}$Fe$_{0.4}$T850 adsorbent, an Hg-TPD experiment was carried out. It is worth mentioning that as a simple and available method Hg-TPD can clarify mercury species adsorbed on solid adsorbents. Figure 7 suggested the Hg-TPD curve of A$_{0.075}$Fe$_{0.4}$T850

![Figure 7. Hg-TPD curves of used A$_{0.075}$Fe$_{0.4}$T850 adsorbent.](https://doi.org/10.1021/acsomega.1c05857)
Hg\(^0\) (ad) + O\(_2\) (ad) → HgO (ad)  
(6)

FeO + \(\frac{1}{2}\)O\(_2\) → FeO\(_3\)  
(7)

3.4. Regeneration Performance of Iron-Modified Biomass Carbon Adsorbents. The regenerability and reusability of used adsorbents are very important in practical industrial applications. Thus, in this study, the recycle tests were conducted on used A\(_{0.075}\)-Fe\(_{0.4}\)-T850 adsorbent. The used A\(_{0.075}\)-Fe\(_{0.4}\)-T850 adsorbent was first heated to 500 °C in N\(_2\) to decompose HgO on the sample. When the Hg\(^0\) concentration at the outlet the online Hg analyzer was lower than 0.3 μg m\(^{-3}\), the removal performance of Hg\(^0\) of the regenerated A\(_{0.075}\)-Fe\(_{0.4}\)-T850 adsorbent could be accessed in N\(_2\)-O\(_2\)-SO\(_2\)-NO-H\(_2\)O-Hg atmosphere at 150 °C. This was considered as a one-cycle process, and the same cycle process was repeated for four times. Figure 8 shows the results. After the first cycle, the removal performance of Hg\(^0\) of regenerated adsorbent remained above 98.6% within 2 h, which was close to that of the fresh sample. The removal performance of Hg\(^0\) of regenerated adsorbent was reduced; however, it was above 92.9%. After four cycles, the removal performance of Hg\(^0\) of the regenerated sample was still more than 85%. These results showed that the A\(_{0.075}\)-Fe\(_{0.4}\)-T850 adsorbent was of great regeneration efficiency, which improved the utilization rate and reduced the cost of the removal of Hg\(^0\).

The magnetic properties of the adsorbent are discussed in section 3.2.3. Herein, the magnetism of the regenerated adsorbent after four cycles was detected. Figure 4b illustrates that the regenerated adsorbent remains as a good superparamagnetic structure after four cycles and its saturation magnetization is lower than that of the fresh sample (25.0 emu g\(^{-1}\)) but still remains at 16.7 emu g\(^{-1}\). Wang et al.\(^{57}\) reported that the saturation magnetization value of magnetic carbon composite derived from pine wood sawdust was 15.6 emu g\(^{-1}\), indicating that the separation of magnetic carbon composite was facile, and almost all adsorbent could be completely recollected using a magnet. Zou et al.\(^{58}\) reported that the improved Fe–Ti spinel remained as a great superparamagnetic structure. The saturation magnetization of it has decreased from 24.6 to 11.8 emu g\(^{-1}\) after the four cycles of Hg\(^0\) capture/recovery. These results indicated that the Fe/BC sample exhibited good magnetization performance, which received assistance from an external magnetic field to separate from fly ash.

4. CONCLUSION

In this study, Fe/BC magnetic adsorbent derived from pepper straw was prepared by a one-step method with K\(_2\)C\(_2\)O\(_4\) and Fe(NO\(_3\))\(_3\) as activator and catalyst precursor, respectively. Its Hg\(^0\) removal efficiency was studied in the fixed-bed system. When the activation temperature was 850 °C, the removal performance of Hg\(^0\) of iron-modified biomass carbon (Fe/BC) adsorbent was excellent, above 97.6% at 150 °C. The Fe/BC showed a wide temperature range for capturing Hg\(^0\). The Hg\(^0\) removal efficiency of Fe/BC adsorbent increased to 100% at 150 °C under N\(_2\) + O\(_2\) + SO\(_2\). Nevertheless, addition of NO resulted in significant inhibition of Hg\(^0\) capture. The Hg\(^0\) removal efficiency of the magnetic Fe/BC adsorbent increased first and then decreased with the increase of the reaction temperature. Regarding the Hg\(^0\) adsorption mechanism, the lattice oxygen in Fe\(_2\)O\(_3\) and chemically adsorbed oxygen as the active site for Hg\(^0\) removal, the Hg species on the used Fe/BC adsorbents was HgO. The results of experiments suggested that after four capture–regeneration cycles Fe/BC adsorbent showed a good regeneration and magnetization performance; these results indicated that pepper straw can be used as a potential raw material for the preparation of biomass carbon adsorbents.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.1c05857.

Effect of NO intermittent supply for Hg\(^0\) removal performance of Fe/BC adsorbent, Raman spectra of Fe/BC adsorbent, the comparison of Hg\(^0\) adsorption capacity between Fe/BC and other biomass AC (PDF)

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Notes

The authors declare no competing financial interest.

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