Effect of Na₂CO₃, HF, and CO₂ Treatment on the Regeneration of Exhausted Activated Carbon Used in Sintering Flue Gas

Jianlu Zheng, Xiangdong Xing,* Zhuogang Pang, Sunxuan Wang, Yueli Du, and Ming Lv*

ABSTRACT: The method of continuous treatment with Na₂CO₃ solution, HF solution, and CO₂ was proposed for the regeneration of the exhausted activated carbon (EAC) produced in the sintering flue gas purification process. In order to obtain the optimal operation conditions, the effect of key parameters such as Na₂CO₃ solution concentration, HF solution concentration, and CO₂ activation temperature on the sulfur conversion rate and regeneration efficiency was analyzed. Also, the N₂ adsorption, Brunauer–Emmett–Teller analysis, scanning electron microscopy-energy dispersive spectrometry, X-ray diffraction, X-ray fluorescence, and Fourier transform infrared spectroscopy were adopted to investigate the deactivation reason and the change of the physical–chemical properties. The results showed that the deactivated EAC was mainly due to the deposition of inorganic compounds such as CaSO₄, SiO₂, and KCl to block the pores. Continuous treatment with Na₂CO₃ solution and HF solution could remove the inorganic compounds effectively. CO₂ activation treatment further developed the blocked porosity and decreased the surface acidity. The optimal conditions for the regeneration of EAC were a Na₂CO₃ concentration of 0.5 mol/L, an HF concentration of 0.8 mol/L, and a CO₂ activation temperature of 1073 K with the activation time of 1 h, corresponding to the specific surface area of 607.91 m²/g. In the fourth regeneration cycle, the adsorption performance during the successive adsorption–regeneration process could still maintain a high level and the regeneration efficiency was 95.31%.

1. INTRODUCTION

Activated carbon (AC) has good adsorption performance due to having a large specific surface area, well-developed pore structure, and abundant functional groups. In recent years, it is increasingly applied in the sintering flue gas purification industry, such as desulfurization. After a period of industrial application, the adsorption capacity of AC could not be recovered by desorption and it is difficult to continue to use, which produces a lot of exhausted activated carbon (EAC). The EAC is usually treated as hazardous waste due to that the adsorbed substance contains sulfur dioxide (SO₂) and dioxins as well as other dangerous pollutants from sintering flue gas. At present, the main way to dispose EAC is incineration or dumping in landfills, which often leads to serious secondary environmental pollution and resource waste. Therefore, the regeneration of EAC has attracted increasing attention recently because of its economic, environmental, and energy benefits.

The regeneration mainly removes the adsorbed pollutants through desorption and decomposition to restore the adsorption capacity of activated carbon. The choice of regeneration technology is closely related to the type and characteristics of adsorbed substances. Research by Guilane and Hamdaoui reported that low-frequency ultrasonication could desorb 4-chlorophenol with a stable structure in EAC from petroleum and pharmaceutical wastewater and the regenerated activated carbon do not differ significantly with...
the virgin carbon. Liu et al. investigated that the magnesium sulfate with a low melting point deposited on Mn-based EAC from coal-fired flue gas is decomposed under 650 °C thermal regeneration and the specific surface area is 800 m²/g.12 Ma et al. found that the Fenton oxidation can degrade hydrocarbon compounds toluene in saturated activated carbon produced in chemical industry waste gas and the regeneration rate of activated carbon is 94.52%.13 A work by Guo et al. reported that the amount of organic pollutant phenol and its derivatives in EAC from coking wastewater can be extracted by the low boiling point organic solvents and the regeneration efficiency is 98.27%.14 However, the EAC produced in the sintering flue gas purification process contains a variety of high thermal stability pollutants such as sulfate, alkali metal, and dust,15 which makes it difficult to regenerate by traditional technology. Up to now, the regeneration technology of EAC from sintering flue gas has not been systematically studied. In order to achieve sustainable development of the steel industry, it is necessary to develop a feasible regeneration method for EAC produced in the sintering flue gas purification process.

In this paper, the continuous treatment method with Na₂CO₃ solution, HF solution, and CO₂ are proposed to regenerate EAC. The deposited sulfate is converted into carbonate and then removed. Also, the texture properties are also changed to enhance the adsorption capacity by CO₂ treatment. The effects of the adsorption—regeneration cycles and regeneration conditions such as Na₂CO₃ solution concentration, HF solution concentration, and CO₂ activation temperature on the sulfur conversion rate and regeneration efficiency are investigated. In addition, the EAC properties are analyzed by N₂ adsorption, Brunauer—Emmett—Teller (BET) analysis, scanning electron microscopy-energy dispersive spectrometry (SEM-EDS), X-ray diffraction (XRD), X-ray fluorescence spectroscopy (XRF), and Fourier transform infrared spectroscopy (FTIR). The regeneration efficiency is evaluated at a fixed bed reactor under simulating sintering flue gas.

2. RESULTS AND DISCUSSION

2.1. Characterization of EAC. The content of C of the FAC and EAC were obtained by ultimate analysis and the other chemical elements contents by XRF analysis, as summarized in Table 1. It could be seen that the C content of EAC was less than that of FAC, as FAC was 93.62 wt %, while the EAC was reduced to 83.06 wt %. The content of S was increased from 0.21 to 4.96 wt %, and the content of Ca, Si, and Cl were 2.21, 1.85, and 1.77 wt %, respectively. This was because the ions adsorbed on the carbon were not eliminated during the sintering flue gas purification process but reacted to form inorganic compounds to deposit on the activated carbon.

Figure 1 presents the XRD pattern of EAC and FAC. The characteristic peaks of both RAC and EAC could be observed at 2θ = 26° and 2θ = 43°, representing the graphite crystallite structure (002) and graphite-like structure (100) or (101) of the carbon materials,16 suggesting that the amorphous and graphite carbon structure of activated carbon were not affected. For FAC, no other characteristic peaks were shown in the XRD pattern, which confirmed its purity. In contrast, the characteristic peaks CaSO₄, SiO₂, and a small amount of KCl appear in the XRD pattern of EAC, which was due to the inorganic compounds deposited during the sintering flue gas purification process. The formation of CaSO₄ might be due to the reaction of SO₂ and Ca-based substances such as CaO in the sintering flue gas purification process. The existence of SiO₂ and KCl was attributed to the deposition of particulates or dust in the sintering flue gas. Also, the CaSO₄, SiO₂, and KCl were difficult to desorb because of their good thermal stability.

To understand the microstructure and the distribution of inorganic compounds deposited on EAC, SEM images of EAC and FAC are shown in Figure 2. It can be seen from Figure 2a—c that the surface of FAC was smooth and the pores were clearly visible. There were also many circular pores with different pore diameters. The surface of EAC was rough, and the pore channel was completely covered by the acicular, lump, and graininess compounds. Combining with the SEM image and EDS analysis, the acicular compounds were CaSO₄, which was mainly distributed in two parts. Most of the CaSO₄ were around the pores, and a small amount was attached to the lump compounds. The phases marked “2” represent the gray lumps embedded in the pores of the activated carbon, which were SiO₂. In addition, a small number of angular particles existed on the surface of the activated carbon, which were identified as KCl. The CaSO₄, SiO₂, and KCl were deposited into the pores, resulting in the increased in mass transfer resistance and decreased in desulfurization capacity.

The N₂ adsorption isotherms of FAC and EAC are illustrated in Figure 3. The shape of the isotherm of FAC was type I according to the IUPAC classification, which was characteristic of microporous materials. When the relative pressure was less than 0.1, the adsorption volume increased rapidly. In contrast, the EAC exhibited a type IV isotherm, indicating the main presence of mesopores. Also, the adsorption capacity of EAC was very low. When the relative pressure was up to 1.0, the adsorption volume of EAC was less than 5 cm³/g, suggesting the loss of adsorption capacity.

Table 1. Chemical Elements of EAC and FAC Analyzed (wt %)

| sample | C   | S   | Si  | Ca  | Fe  | Cl  | K   | others |
|--------|-----|-----|-----|-----|-----|-----|-----|--------|
| FAC    | 93.62 | 0.21 | 0.14 | 0.16 | 0.07 | 0.11 | 0.06 | 5.63   |
| EAC    | 83.06 | 4.96 | 1.85 | 2.21 | 1.54 | 1.77 | 1.15 | 3.46   |

Figure 1. XRD pattern of EAC and FAC.
The physical properties of FAC and EAC are listed in Table 2. It could be found that the specific surface area seriously reduced from 537 to 2.03 m²/g, while total pore volume dropped from 0.364 to 0.006 cm³/g, implying that the physical properties of EAC significantly deteriorated. The micropore volume was dropped from 0.195 to 0.001 cm³/g, and the average pore size increased from 2.84 nm to 11.65 nm, showing that the number of micropores sharply declined, which led to the increase in the number of mesopores. This had an adverse effect on activated carbon desulfurization because the micropores were the main site of the desulfurization reaction.

2.2. Effect of Na₂CO₃ Concentration on Sulfur Conversion Rate of EAC

Na₂CO₃ solution was used to convert the CaSO₄ into CaCO₃ and removed the water-soluble compounds such as KCl. The change of sulfur content on the EAC was defined as the sulfur conversion rate, which was calculated through eq 1,

\[
\text{sulfur conversion rate} = \left(\frac{M_0 \times W_0 - M_1 \times W_1}{M_0 \times W_0}\right) \times 100\%
\]

where \(M_0\) refers to the mass of EAC, g; \(W_0\) refers to the mass percentage of S on EAC, wt %; \(M_1\) and \(W_1\) refer to the mass of EAC treated by Na₂CO₃ solution, g; and \(W_1\) refer to the mass percentage of S on EAC treated by Na₂CO₃ solution, wt %.

The relationship between the Na₂CO₃ solution concentration and sulfur conversion rate is shown in Figure 4. It could be concluded that at low solution concentration of Na₂CO₃, the increased in solution concentration also increased the sulfur conversion rate. When the solution concentration of Na₂CO₃ was 0.1 mol/L, the sulfur conversion rate was only 75.02%. As the solution concentration continued to rise to 0.5 mol/L, the sulfur conversion rate was as high as 94.60%. This was because the diffusion rate of CO₃²⁻ in the solution was accelerated with the increase of solution concentration to better contact with sulfate, leading to an enhanced conversion effect, as expressed in eq 2. However, When the Na₂CO₃ concentration exceeded 0.5 mol/L, the change of sulfur conversion rate slowed down, which was determined as the optimum Na₂CO₃ concentration.

\[
\text{CaSO}_4 + \text{Na}_2\text{CO}_3 = \text{CaCO}_3 + \text{Na}_2\text{SO}_4
\]
The phases were mainly CaCO₃ and SiO₂. The characterization peaks of CaSO₄ and KCl disappeared, which indicated that the sulfur conversion efficiency was high. The CaCO₃ was identified as conversion product, and the KCl was washed by deionized water. SiO₂ had no change in the reaction during the conversion process.

The surface morphology and EDS analysis of EAC after Na₂CO₃ solution treatment under different magnifications are shown in Figure 6. The pore structure of EAC could not be observed clearly, and the shapes of impurity compounds were lumps and flakes corresponding to the CaCO₃ and SiO₂, respectively. The CaCO₃ was latched onto the pores, and the SiO₂ was distributed around the pore.

### 2.3. Effect of HF Concentration on Regeneration Efficiency of EAC

HF solution treatment was carried out to improve the desulfurization capacity by removing the inorganic compounds deposited on activated carbon. However, the degree of regeneration was greatly dependent on the treatment conditions. The effect of HF solution concentration on the regeneration efficiency is shown in Figure 7. It could be found that the regeneration efficiency first increased significantly. The regeneration efficiency reached the maximum value of 81.55% when the HF concentration was 0.8 mol/L due to the reaction shown in eqs 3 and 4:

\[
\text{CaCO}_3 + 2\text{HF} = \text{CaF}_2 + \text{H}_2\text{O} + \text{CO}_2
\]  

(3)

\[
\text{SiO}_2 + 4\text{HF} = \text{SiF}_4 + \text{H}_2\text{O}
\]  

(4)

However, the regeneration efficiency decreased gradually as the HF solution concentration continued to rise. When the solution concentration of HF was 1.4 mol/L, the regeneration efficiency was reduced to 75.02%. This was attributed to two possible features of HF solution treatment. The main HF solution could effectively remove the inorganic compounds. On the other hand, the HF solution had an etching to the surface pore morphology of material. As the HF solution concentration continued to increase, the activated carbon framework was corroded, which caused the original pore structure to collapse. This was because when the concentration increased to a certain extent, HF could react with ash from the attached wall of the activated carbon such as Al, Ca, K, and other metal oxides, causing the change in the pore structure. In addition, the SiF₄ gas generated by the reaction of HF and SiO₂ was not easy to diffuse in the micropores, leading to the collapses of the micropores, which made the micropores expand into mesopores or macropores of the activated carbon.

Figure 8a shows the XRD pattern of RAC-H-0.8. The characterization peaks of CaCO₃ and SiO₂ disappeared. It indicated that the HF solution could effectively remove CaCO₃ and SiO₂, which was conducive to the recovery of the adsorption capacity.

The N₂ adsorption isotherm of RAC-H-0.8 is presented in Figure 8b. The adsorption capacity increased rapidly when the relative pressure was low and the isotherm type was the same as that of FAC. It means that the RAC micropores were well restored after HF treatment. Table 3 lists the physical properties of RAC-H-0.8, showing that the specific surface area, total pore volume, and micropore volume of EAC were 432.14m²/g, 0.274cm³/g, and 0.114cm³/g, respectively, which were close to 80% of the FAC. HF treatment effectively improved the physical properties of EAC.

Figure 9 shows the microstructure of RAC-H-0.8. It was noticed that the pore was irregular and visible. The impurities on the surface of activated carbon after HF solution treatment were not observed.

### 2.4. Effect of Activation Temperature on Regeneration Efficiency of EAC

The RAC had good adsorption performance after Na₂CO₃ solution and HF solution treatment. However, the recovery of texture properties was not significant, and the percentages of microporosity for the total porosity of the EAC were less than that of the FAC. CO₂ activation treatment could improve the texture characteristics of RAC and increase the desulfurization capacity. The effect of activation temperature on the regeneration efficiency and yield of RAC are presented in Figure 10. As the activation temperature increased from 773 to 1073 K, the regeneration efficiency increased from 82.15 to 110.5%, and the yield gradually decreased. This was because the activated carbon reacted with CO₂ at high temperature to reactivate the pristine carbon, which further developed the blocked porosity for EAC and produced new pores. However, the regeneration efficiency reduced seriously to 97.35% when the temperature was 1173 K. It might be due to the fact that the continued enhancement of activation reaction would lead to the original pore structure collapsing, which would lead to the original pore structure to collapse.
being destroyed, and a part of micropores collapsed into mesopores.

The N₂ adsorption isotherm measurements of RAC-C are given in Figure 11a. For the sample after CO₂ activation treatment, the shape of isotherms belonged to type I. The adsorption isotherm of RAC tended to be saturated when the relative pressure was lower than 0.1, which belonged to the characteristics of microporous materials. In addition, the isotherms had a larger slope, indicating that there was a certain amount of mesopores in the RAC when the relative pressure was higher than 0.1.

Table 4 summarizes the physical properties of RAC. The specific surface, total pore volume, and micropore volume continuously increased, but the average pore size gradually reduced with the increase in activation temperature. When the activation temperature was 1073 K, the specific surface area, the micropore volume, and the total pore volume reached the maximum values of 607.91 m²/g, 0.211 cm³/g, and 0.406 cm³/g, respectively. Also, the RAC-C-1073 exhibited better adsorption ability than that of other RAC, even higher than that of FAC. However, the micropore volume reduced rapidly to 0.189 cm³/g, but the total pore increased to 0.433 cm³/g as the temperature increased to 1173 K. It indicated that the RAC contained a large number of mesopores, and the decrease of micropores lead to the relative increase in the average pore size. The CO₂ treatment could effectively improve the physical properties, and the optimal regeneration temperature of EAC was 1073 K.

The FT-IR spectra of RAC after CO₂ activation treatment is provided in Figure 11b. It could be found from Figure 11b that the positions of the stretching vibration peaks were similar except the intensities, which indicated that the type of surface functional groups was consistent. In all the recorded spectra, the adsorption band around 1640 cm⁻¹ could be assigned to the stretching vibration of the C≡C bond on the surface of olefins and the C≡O stretching vibration in carbonyl groups almost did not change after CO₂ activation treatment.21,22 Also, peaks in the 2250−2400 cm⁻¹ range were observed due to the fact that the band of C−O stretching vibration and the C−O bands became a little weaker as the temperature increased.23 Moreover, the stretching vibration of the O−H bands in the 3300−3600 cm⁻¹ range decreased significantly, which was associated with the hydroxyl groups, and carboxyl acid groups as well as chemisorbed water on the surface of RAC decreased.24,25 The functional groups such as C−O bands and O−H bands decreased, which led to the attenuated surface acidity and the release of activating site, which further enhanced the SO₂ adsorption capacity.26 It was also an important reason for the increase of regeneration efficiency after CO₂ activation treatment.

The FT-IR spectra of RAC after CO₂ activation treatment is provided in Figure 11b. It could be found from Figure 11b that the positions of the stretching vibration peaks were similar except the intensities, which indicated that the type of surface functional groups was consistent. In all the recorded spectra, the adsorption band around 1640 cm⁻¹ could be assigned to the stretching vibration of the C≡C bond on the surface of olefins and the C≡O stretching vibration in carbonyl groups almost did not change after CO₂ activation treatment.21,22 Also, peaks in the 2250−2400 cm⁻¹ range were observed due to the fact that the band of C−O stretching vibration and the C−O bands became a little weaker as the temperature increased.23 Moreover, the stretching vibration of the O−H bands in the 3300−3600 cm⁻¹ range decreased significantly, which was associated with the hydroxyl groups, and carboxyl acid groups as well as chemisorbed water on the surface of RAC decreased.24,25 The functional groups such as C−O bands and O−H bands decreased, which led to the attenuated surface acidity and the release of activating site, which further enhanced the SO₂ adsorption capacity.26 It was also an important reason for the increase of regeneration efficiency after CO₂ activation treatment.

The surface morphology of RAC and RAC-H are shown in Figure 12. It could be seen that there were no obvious differences between the morphology of RAC-H and RAC-C.
The surface of RAC was smooth, and the pores were present as a regular circle. As the activation temperature reached 1173 K, the pore size of RAC was obviously larger than that treated under other temperatures.

In order to study the change in regeneration performance with adsorption–regeneration cycles, the effects of the adsorption–regeneration cycle on the regeneration efficiency of activated carbon were carried, and the results are shown in Figure 13. It was obvious that under the optimal experimental conditions used, the adsorption performance during the successive adsorption–regeneration process could still maintain a high level. In the fourth regeneration cycle, the regeneration efficiency was 95.31%. The results showed that the regenerative effect of continuous treatment with Na$_2$CO$_3$ solution, HF solution, and CO$_2$ regeneration was remarkable.

### 3. CONCLUSIONS

A novel continuous treatment method with Na$_2$CO$_3$ solution, HF solution, and CO$_2$ could effectively regenerate EAC from the sintering flue gas purification process. When the Na$_2$CO$_3$ concentration was 0.5 mol/L, HF concentration was 0.8 mol/L, and CO$_2$ activation temperature was 1073 K, the regeneration efficiency was up to 110.5%. The deposition of inorganic compounds such as CaSO$_4$ and SiO$_2$ as well as KCl leads to the deactivation of activated carbon. Na$_2$CO$_3$ solution could convert the CaSO$_4$ into CaCO$_3$ and remove the water-soluble compounds such as KCl. The sulfur conversion rate first increased significantly, and then the change slowed down with the increase in Na$_2$CO$_3$ concentration. When the Na$_2$CO$_3$ concentration was 0.5 mol/L, the sulfur conversion rate radical was up to 94.6%. Also, the CaCO$_3$ was identified as the conversion product. Secondary chemical regeneration using

| Sample   | Specific Surface Area (m$^2$/g) | Total Pore Volume (cm$^3$/g) | Micropore Volume (cm$^3$/g) | Average Pore Size (nm) |
|----------|---------------------------------|-----------------------------|-----------------------------|------------------------|
| RAC-H-0.8| 432.14                          | 0.274                       | 0.114                       | 4.56                   |

Figure 8. Structure characteristics of RAC-H-0.8: (a) XRD pattern; (b) N$_2$ adsorption isotherm.

Table 3. Physical Properties of RAC-H-0.8

Figure 9. SEM images of RAC-H-0.8 under different magnifications: (a) 1.0 k× and (b) 3.0 k×.

Figure 10. Effect of activation temperature on regeneration efficiency and yield.
HF solution after the Na\textsubscript{2}CO\textsubscript{3} solution treatment removed residual inorganic compounds, and the specific surface area of the activated carbon recovered to 80% of FAC. The regeneration efficiency increased at first and then decreased with the increase in HF solution concentration. Finally, CO\textsubscript{2} activation treatment developed the blocked porosity and reduced the surface acidity, further improving the adsorption performance of activated carbon. In the four adsorption–regeneration processes, the adsorption performance could still maintain a high level.

4. EXPERIMENTAL SECTION

4.1. Materials and Equipment. The EAC was derived from the steel plant in China. The solutions used in the experiment were prepared with analytical grade Na\textsubscript{2}CO\textsubscript{3} (Damao Chemical Factory, Tianjin, China) and HF solution (Damao Chemical Factory, Tianjin, China, 40%). All aqueous solutions were prepared with deionized water. The numerical control ultrasonic instrument (model no. KQ5200DE, Ultrasonic instrument Co., Ltd., Kunshan, China) was used to produce ultrasound for the regeneration of EAC. CO\textsubscript{2} activation treatment was carried out in a vacuum tubular}

| sample   | specific surface area (m\textsuperscript{2}/g) | total pore volume (cm\textsuperscript{3}/g) | micropore volume (cm\textsuperscript{3}/g) | average pore size (nm) |
|----------|-----------------------------------------------|--------------------------------------------|-------------------------------------------|------------------------|
| RAC-H-0.8| 432.14                                        | 0.274                                      | 0.114                                     | 4.56                   |
| RAC-C-773| 447.36                                        | 0.323                                      | 0.165                                     | 3.98                   |
| RAC-C-873| 464.69                                        | 0.328                                      | 0.186                                     | 3.95                   |
| RAC-C-973| 517.32                                        | 0.345                                      | 0.190                                     | 3.94                   |
| RAC-C-1073| 607.91                                      | 0.406                                      | 0.211                                     | 3.91                   |
| RAC-C-1173| 529.45                                       | 0.433                                      | 0.189                                     | 4.12                   |

Figure 11. Structure characteristics of RAC-C: (a) N\textsubscript{2} adsorption isotherms of RAC-C; (b) FT-IR spectra of RAC-C.

Figure 12. SEM images of the (a) RAC-H-0.8, (b) RAC-C-773, (c) RAC-C-873, (d) RAC-C-973, (e) RAC-C-1073, and (f) RAC-C-1173.

Table 4. Physical Properties of RAC

![Image](https://doi.org/10.1021/acsomega.1c04182)

ACS Omega 2021, 6, 25762−25771
furnace (Jinbi Electrical Equipment Co., Ltd., Chongqing, China).

4.2. Experimental Methods. The schematic diagram of the experimental flow chart is shown in Figure 14. The EAC was washed in deionized water and dried at 378 K for 24 h, which would be used in the following regeneration process. First, 10 g of EAC was added to the beaker containing 100 mL Na₂CO₃ solution with different concentrations of 0.1, 0.2, 0.3, 0.4, 0.5, and 0.6 mol/L for pretreatment and then placed in the ultrasonic instrument with the ultrasound power of 100 W and reaction temperature of 333 K. Once the reaction time was up to 1 h, the EAC was taken out to be washed with deionized water until the washing solution reached neutral and part of sample was used to analyze the properties after drying for 24 h at 378 K.

Second, 5 g of the EAC after the Na₂CO₃ solution treatment was immersed in the beaker containing 50 mL HF solution with different concentrations of 0.6, 0.8, 1.0, 1.2, and 1.4 mol/L and stirred at room temperature. After the HF solution treatment for 30 min, the samples were rinsed with deionized water for 1 h and part of sample was used to analyze the properties after drying for 24 h at 378 K.

Finally, 5 g of the EAC treated by Na₂CO₃ solution and HF solution was loaded into the vacuum tubular furnace with the temperature increasing at a constant rate of 7 K/min to the set target temperature under a N₂ atmosphere, which was shifted to CO₂ and reacted for 1 h when the set temperature was reached with the gas flow rate of 40 mL/min. The nomenclature for the applied regeneration methods is summarized in Table 5.

4.3. Regeneration Efficiency Testing. The regeneration efficiency of activated carbon was evaluated in the fixed bed reactor, and the equipment apparatus is shown in Figure 15. The equipment was mainly consisted of a gas mixing system, adsorption reaction system, and gas analysis system. The reactor had a 10 mm inner diameter and was 200 mm high. A total of 1 g activated carbon was put into the reactor before testing. The composition of the mixture gas based on the actual sintering flue gas contained 1200 ppm SO₂, 10 vol % H₂O, 15 vol % O₂, and N₂ as balance. The flue gas analyzer was used to monitor the SO₂ concentration, and the reaction temperature was 393 K. The regeneration efficiency of RAC was calculated by the SO₂ adsorption capacity of RAC and FAC, as shown in eq 5:

\[ R_e = \frac{R_r}{R_0} \times 100\% \]  

where \( R_e \) refers to the regeneration efficiency; \( R_r \) and \( R_0 \) refer to the adsorption capacity of RAC and fresh activated carbon (FAC), respectively.

4.4. Characterization. \( \text{N}_2 \) adsorption–desorption experiment was conducted to characterize the pore structure of activated carbon on an aperture analyzer (V-Sorb2800P, Kine Spectrum technology, China) at 77 K. The specific surface area and total pore volume were calculated by the Brunauer–Emmett–Teller (BET) equation, and the micropore volume was obtained by the \( T \)-plot method. The phase composition of the inorganic compounds deposited was analyzed by X-ray diffraction (XRD, D8 ADVANCE A25, Bruker AXS, Germany) using Cu Kα radiation (\( \lambda = 1.5406 \) Å), and the measuring method was stated in our previous study.²⁷ Fourier transform infrared spectroscopy (FT-IR, Nicolet 6700, Thermo Fisher Scientific, USA) was used to characterize the surface functional groups. The 2.0 mg RAC samples with 400 mg potassium bromide (KBr) were mixed and pressured into the thin film. The wavenumber range of RAC was recorded at a scope of 400–4000 cm⁻¹, and the scanning time and resolution were set as 32 s and 4 cm⁻¹, respectively.

Figure 13. Effect of regeneration efficiency in successive adsorption–regeneration cycles.

Figure 14. Schematic diagram of the experimental flow chart.

| Table 5. Nomenclature of RAC³⁴ |
|--------------------------------|
| sample name | Na₂CO₃ solution treatment | HF solution treatment | CO₂ treatment |
| RAC-Y-S | yes | no | no |
| RAC-H-S | yes | yes | no |
| RAC-C-T | yes | yes | yes |

\( ^{\text{a}} \) Y refers to the pretreatment method of Na₂CO₃ solution; S refers to the solution concentration; H refers to the treatment method with HF solution; C refers to the CO₂ activation treatment; and T refers to the regeneration temperature.
X-ray fluorescence (XRF, S4 Explorer, Bruker AXS, Germany) was used to analyze the content of the chemical elements of the EAC and FAC. The samples were ground to −0.074 μm.

The surface morphology of samples was observed by using a scanning electron microscope (SEM, Quanta200, FEI Company, USA) with an acceleration voltage of 5 kV. The elemental composition was analyzed by energy-dispersive spectroscopy (EDS, Oxford instruments company, U.K.).

**AUTHOR INFORMATION**

**Corresponding Authors**

Xiangdong Xing — School of Metallurgical Engineering, Xi’an University of Architecture and Technology, Xi’an 710055, PR China; Metallurgical Engineering Technology Research Center of Shaanxi Province, Xi’an 710055, PR China; orcid.org/0000-0002-1721-5046; Email: xaxxd@xauat.edu.cn

Ming Lv — School of Metallurgical Engineering, Xi’an University of Architecture and Technology, Xi’an 710055, PR China; Metallurgical Engineering Technology Research Center of Shaanxi Province, Xi’an 710055, PR China; Email: 799636613@qq.com

**Authors**

Jianlu Zheng — School of Metallurgical Engineering, Xi’an University of Architecture and Technology, Xi’an 710055, PR China; Metallurgical Engineering Technology Research Center of Shaanxi Province, Xi’an 710055, PR China

Zhuogang Pang — School of Metallurgical Engineering, Xi’an University of Architecture and Technology, Xi’an 710055, PR China; Metallurgical Engineering Technology Research Center of Shaanxi Province, Xi’an 710055, PR China

Sunxuan Wang — Shaanxi Institute for Food and Drug Control, Xi’an 710065, PR China

Yueli Du — School of Metallurgical Engineering, Xi’an University of Architecture and Technology, Xi’an 710055, PR China; Metallurgical Engineering Technology Research Center of Shaanxi Province, Xi’an 710055, PR China

Complete contact information is available at:

https://pubs.acs.org/10.1021/acsomega.1c04182

**Notes**

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

The present work was financially supported by the Natural Science Basic Foundation of China (program no. 51874058) and China Postdoctoral Science Foundation (grant no. 2019M663932XB). The authors gratefully acknowledge their support.

**REFERENCES**

(1) Xing, B.; Zeng, H.; Huang, G.; Jia, J.; Yuan, R.; Zhang, C.; Sun, Q.; Cao, Y.; Chen, Z.; Liu, B. Magnesium citrate induced growth of noodle-like porous graphitic carbons from coal tar pitch for high-performance lithium-ion batteries. *Electrochim. Acta* 2021, 376, 138043.

(2) Li, Q.; Qi, L.; Gao, C. Chemical regeneration of spent powdered activated carbon used in decolorization of sodium salicylate for the pharmaceutical industry. *J. Cleaner Prod.* 2015, 86, 424–431.

(3) Wang, Z.; Xing, B.; Zeng, H.; Huang, G.; Liu, X.; Guo, H.; Zhang, C.; Cao, Y.; Chen, Z. Space-confined carbonization strategy for synthesis of carbon nanosheets from glucose and coal tar pitch for high-performance lithium-ion batteries. *Appl. Surf. Sci.* 2021, 547, 149228.

(4) Guilane, S.; Hamdaoui, O. Ultrasound-assisted regeneration of granular activated carbon saturated by 4-chlorophenol in batch-loop reactor. *Desalin. Water Treat.* 2015, 57, 17262–17270.

(5) Liu, S.; Wang, Y.; Wang, B.; Jun, H.; Deng, S.; Yu, G. Regeneration of Rhodamine B saturated activated carbon by an electro-peroxone process. *J. Cleaner Prod.* 2017, 168, 584–594.

(6) Nahm, S. W.; Shim, W. G.; Park, Y. K.; Kim, S. C. Thermal and chemical regeneration of spent activated carbon and its adsorption property for toluene. *Chem. Eng. J.* 2012, 210, 500–509.

(7) Miguez, M.; Goetz, V.; Plantard, G.; Jaeger, Y. Sustainable Thermal Regeneration of Spent Activated Carbons by Solar Energy: Application to Water Treatment. *Ind. Eng. Chem. Res.* 2016, 55, 7003–7011.

(8) Cheng, J.-Y.; Yang, L.; Dong, L.; L, X.-j.; Yuan, W.-k. Regeneration of hexamminecobalt(II) catalyzed by activated carbon treated with KOH solutions. *J. Hazard. Mater.* 2011, 191, 184–189.
(9) Yang, L.; Yao, L.; Jiang, W.; Jiang, X.; Li, J. The study on continuous denitrification, desulfurization of pyrolusite/activated coke hybrid catalyst. RSC Adv. 2018, 8, 406–413.

(10) Qiao, K.; Yu, J.; Zhu, B.; Chi, C.; Di, C.; Cheng, Y.; Shang, M.; Li, C. Oxygen-Rich Activated Carbon Fibers with Exceptional Cu(II) Adsorptivity and Recycling Performance. Ind. Eng. Chem. Res. 2020, 59, 13088–13094.

(11) Zhou, L.; Liao, C.; Li, T.; An, J.; Du, Q.; Wana, L.; Li, N.; Pan, X.; Wang, X. Regeneration of activated carbon air-cathodes by half-wave rectified alternating fields in microbial fuel cells. Appl. Energy 2018, 219, 199–206.

(12) Liu, Y.-J.; Qu, Y.-F.; Guo, J.-X.; Wang, X.-J.; Chu, Y.-H.; Yin, H.-Q.; Li, J.-J. Thermal Regeneration of Manganese Supported on Activated Carbons Treated by HNO3 for Desulfurization. Energy Fuels 2015, 29, 1931–1940.

(13) Ma, L.; Wu, Z.; He, M.; Wang, L.; Yang, X.; Wang, J. Experimental study on Fenton oxidation regeneration of adsorbed toluene saturated activated carbon. Environ. Technol. 2020, 1–10.

(14) Guo, D.; Shi, Q.; He, B.; Yuan, X. Different solvents for the regeneration of the exhausted activated carbon used in the treatment of coking wastewater. J. Hazard. Mater. 2011, 186, 1788–1793.

(15) Guo, J.; Li, Y.; Xiong, J.; Zhu, T. Coupling mechanism of activated carbon mixed with dust for flue gas desulfurization and denitrification. J. Environ. Sci. 2020, 98, 205–214.

(16) Shi, Q.; Ding, L.; Long, H.-M.; Chun, T.-J. Study of Catalytic Combustion of Dioxins on Ce-V-Ti Catalysts Modified by Graphene Oxide in Simulating Iron Ore Sintering Flue Gas. Materials 2020, 13, 125.

(17) Xing, X.; Du, Y.; Zheng, J.; Wang, S.; Ren, S.; Ju, J. Isothermal Carbothermal Reduction of FeTiO3 Doped with MgO. JOM 2021, 73, 1328.

(18) Zhou, Z.; Huang, T.; Yu, A. Three-Dimensional Flower-Shaped Activated Porous Carbon/Sulfur Composites as Cathode Materials for Lithium–Sulfur Batteries. ACS Sustainable Chem. Eng. 2014, 2, 2442–2447.

(19) Lv, M.; Li, D.; Zhang, Z.; Logan, B. E.; Liu, G.; Sun, M.; Dai, C.; Feng, Y. Unveiling the correlation of Fe3O4 fractions upon the adsorption behavior of sulfamethoxazole on magnetic activated carbon. Sci. Total Environ. 2021, 757, 143717.

(20) Zeydanli, D.; Akman, S.; Vakifahmetoglu, C. Polymer-derived ceramic adsorbent for pollutant removal from water. J. Am. Ceram. Soc. 2018, 101, 2258–2265.

(21) Saleh, T. A.; Sarı, A.; Tuzen, M. Optimization of parameters with experimental design for the adsorption of mercury using polyethyleneimine modified-activated carbon. J. Environ. Chem. Eng. 2017, 5, 1079–1088.

(22) Feng, P.; Li, J.; Wang, H.; Xu, Z. Biomass-Based Activated Carbon and Activators: Preparation of Activated Carbon from Corn cob by Chemical Activation with Biomass Pyrolysis Liquids. ACS Omega 2020, 5, 24064–24072.

(23) Ledesma, B.; Román, S.; Álvarez-Murillo, A.; Sabio, E.; González, J. F. Cyclic adsorption/thermal regeneration of activated carbons. J. Anal. Appl. Pyrolysis 2014, 106, 112–117.

(24) MacDermid-Watts, K.; Adewakun, E.; Norouzi, O.; Abhi, T. D.; Pradhan, R.; Dutta, A. Effects of FeCl3 Catalytic Hydrothermal Carbonization on Chemical Activation of Corn Wet Distillers’ Fiber. ACS Omega 2021, 6, 14875–14886.

(25) Liu, X.; Wang, J. Electro-assisted adsorption of Cs(I) and Co(II) from aqueous solution by capacitive deionization with activated carbon cloth/graphene oxide composite electrode. Sci. Total Environ. 2020, 749, 141524.

(26) Li, H.; Han, X.; Huang, H.; Wang, Y.; Zhao, L.; Cao, L.; Shen, B.; Gao, J.; Xu, C. Competitive adsorption desulfurization performance over K-Doped NiY zeolite. J. Colloids Interface Sci. 2016, 483, 102–108.

(27) Xing, X.; Pang, Z.; Chuan, M.; Wang, S.; Ju, J. Effect of MgO and BaO on viscosity and structure of blast furnace slag. J. Non-Cryst. Solids 2020, 530, 119801.