

Research Article

Kinetics of SO₂ Adsorption on Powder Activated Carbon in a Drop Tube Furnace

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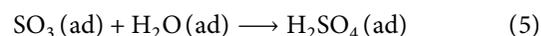
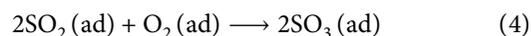
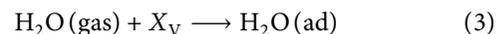
SO₂ (S) adsorption kinetics on the powdered activated carbons (AC) was performed in a laboratory-scale drop tube furnace. The reaction conditions affecting the adsorption of SO₂, such as AC/S molar ratio, temperature, the concentration of SO₂, O₂, and H₂O, and AC circulation rate, were studied. The powdered AC rapidly adsorbs SO₂ in the initial 1.2 seconds and then the amount of SO₂ adsorption slowly increases. The SO₂ removal rate increases with the increasing of AC/S molar ratio, the decreasing of adsorption temperature, and inlet SO₂ concentration. The O₂ and H₂O are beneficial to SO₂ removal by powdered AC. SO₂ removal rate drops to 6.83% after 14 cyclic adsorptions. The powdered AC circulation increases SO₂ removal rate from 62.35% to 99.42% with AC circulation ratio = 7. The SO₂ adsorption kinetics can be predicted by Bangham model.

1. Introduction

Emission of sulfur dioxide (SO₂) has resulted in worldwide concerns in recent decades [1–3]. Adsorptive flue gas desulfurization (FGD) by activated carbon (AC) has been utilized in various industrial processes due to the advantage in recycling SO₂ as sulfuric acid or sulfur and combined removal of other hazardous gasses [1–7]. Conventional adsorptive FGD uses pelletized AC with 5–9 mm diameter as adsorbent in the moving bed reactor. Efficient removal of SO₂ is achievable with this method. However, high construction investment and operating consumption have always been a concern in some commercialization. The costs are mainly derived from the consumption of AC, which

further results from the low utilization of inner part of pelletized AC and abrasion [8–10]. The powdered AC with diameter less than 100 μm, that was expected to have higher utilization ratio and lower production cost compared with the pelletized AC, is selected as the adsorbent in a novel method proposed by Ma [8, 11, 12]. The circulating fluidized bed (CFB) reactor is designed in the new method.

As for SO₂ adsorption fundamentals over AC, the following processes have been widely accepted with researchers [7]. The SO₂ in flue gas is first adsorbed on the surface of AC and then catalyzed to sulfuric acid (H₂SO₄) and H₂SO₄ is stored in pores of AC. The relevant reactions involved are as follows [7]:



where (gas) is for gas phase, X_V denotes the vacant sites, and (ad) represents the adsorbed species.

According to the adsorption mechanism above, the powdered AC would certainly show more rapid SO₂ adsorption rate compared with the pelletized AC due to larger surface area exposed to the gas and shorter diffusion distance

for adsorption process. When such powdered AC is utilized in a CFB reactor, the adsorption fundamentals are different with the pelletized AC in moving bed. These adsorption fundamentals over the powdered AC in fluidized bed reactor, especially for the adsorption dynamics, are required before further development of the new method.

The relative amount of SO₂ and AC in the gas flow is an important value that would affect the adsorption dynamics. In order to simplify the value, the AC-SO₂ (AC/S) molar ratio is proposed in this paper. With the assumption that the molar mass of activated carbon is 12 g/mol, AC/S molar ratio is defined as the molar ratio of AC to SO₂.

Aimed at studying the adsorption dynamics in CFB, continuous supply of simulated flue gas and powdered AC in the experiment is essential. A drop tube reactor which can supply stable AC/S molar ratio with easy access as bench scale study is designed to simulate the adsorption process in CFB reactor.

In the paper, the SO₂ adsorption kinetics on powdered AC is performed in a laboratory-scale drop tube furnace. The reaction conditions affecting the adsorption of SO₂, such as AC/S molar ratio, temperature, the concentration of SO₂, O₂, and H₂O, and AC circulation rate, are studied. The SO₂ adsorption kinetics model is also proposed.

2. Experiments and Methods

2.1. Activated Carbon. A granular AC, which was made from coal by Shanghai Activated Carbon Co., Ltd., was used as raw material. The granular ACs were ground and screened and then 0.075 mm powdered ACs were selected as the research object. The N₂ adsorption isotherms of AC were measured at 77 K by a Micromeritics ASAP2020 instrument. The Brunauer-Emmett-Teller (BET) equation and *t*-plot method were used to calculate the specific surface area (*S*_{BET}) and the micropore specific surface area (*S*_{mic}) of AC, respectively. The *t*-plot method and the Barrett-Joyner-Halenda (BJH) method were used to calculate the micropore volume (*V*_{mic}) and mesopore volume (*V*_{mes}), respectively. The Horvath-Kawazoe (HK) method was used to calculate the micropore size (*L*_{mic}). The HK method and BJH method were used to calculate the size distribution of micropore and mesopore, respectively.

The Fourier transformed infrared spectroscopy (FT-IR) spectra of the powdered AC was measured by Vertex-70 from Brook Company in Germany. The spectrum was recorded from 4000 to 400 cm⁻¹ at a resolution of 4 cm⁻¹.

2.2. SO₂ Adsorption. The SO₂ adsorption kinetics on the powdered AC was performed in a laboratory-scale drop tube furnace, and the system diagram of the drop tube furnace is shown in Figure 1. The stainless steel reactor of the drop tube furnace is 2 meters in length and 40 mm in diameter. The reaction temperature is controlled by an electric heating device. There are five gas sampling holes, which were evenly arranged along the reactor, to measure SO₂ concentration. The MFEV-1VO microfeeder continuously and uniformly delivers the powdered AC into the reactor.

SO₂, O₂, H₂O, and N₂ constitute the simulated flue gas and the concentration of SO₂, O₂, and N₂ is controlled by mass flowmeters (Sevenstar CS200). The peristaltic pump transports deionized water to the evaporator to generate water steam. The electric heating device heats the simulated flue gas to the reaction temperature to avoid condensation of water vapor before entering the reactor. The flow rate of the simulated flue gas is 20 L/min. The SO₂ concentration is between 0.015% and 0.1%, the O₂ concentration is between 0% and 6%, the H₂O concentration is between 0% and 8%, and the rest is N₂. The reaction temperature is between 65°C and 95°C.

The FT-IR gas analyzer (Gasetm Dx4000) was used to measure the SO₂ concentration in flue gas. The SO₂ removal rate by the powdered AC was determined by the following equation:

$$\eta = \frac{C_{\text{SO}_2\text{-in}} - C_{\text{SO}_2\text{-out}}}{C_{\text{SO}_2\text{-in}}} \times 100\%, \quad (6)$$

where η is desulfurization efficiency, $C_{\text{SO}_2\text{-in}}$ is the SO₂ concentration at the reactor inlet, and $C_{\text{SO}_2\text{-out}}$ is the SO₂ concentration at the sampling port.

The adsorption amount of SO₂ on the powdered AC was determined by the following equation:

$$q = 28.6 \frac{C_{\text{SO}_2\text{-in}} V \eta}{m_{\text{AC}}}, \quad (7)$$

where q is the adsorption amount of SO₂ on the powdered AC, mg/g, V is the flue gas flow rate, L/min, and m_{AC} is the feeding rate of the powdered AC, g/min.

3. Results and Discussions

3.1. Characterization of the Powdered AC. The N₂ adsorption isotherms of the granular AC and powdered AC are shown in Figure 2, the shape of which is type I according to IUPAC, indicating that the powdered AC mainly contains micropores [13].

Table 1 shows the pore structure parameters of the granular AC and powdered AC. Figure 3 shows the pore-size distribution of the granular AC and powdered AC. The value of *S*_{mic} of the powdered AC is 541 m²/g, which is larger than that of granular AC. The value of *L*_{mic} of the powdered AC is 0.699 nm, which is larger than the molecular dynamics diameter of SO₂, O₂, and H₂O, meaning that SO₂, O₂, and H₂O can diffuse into the micropores of the powdered AC [8].

Because the particle mass transfer and pore diffusional resistances of the granular AC are larger than those of the powdered AC, N₂ molecules cannot diffuse into some micropores inside the granular AC; therefore, the values of *S*_{BET}, *S*_{mic}, and *V*_{mic} of the granular AC are lower than that of the powdered AC.

The FT-IR spectrum of the powdered AC is shown in Figure 4. The sample had adsorption bands at around 1560, 1414, and 1107 cm⁻¹. The one at 1560 cm⁻¹ could be associated with quinone and carbonyl groups. The 1414 cm⁻¹ can be attributed either to carboxyl groups or to phenolic hydroxyl groups. The adsorption band at 1107 cm⁻¹ is related to C–O stretching in ethers, lactones, and phenols [8].

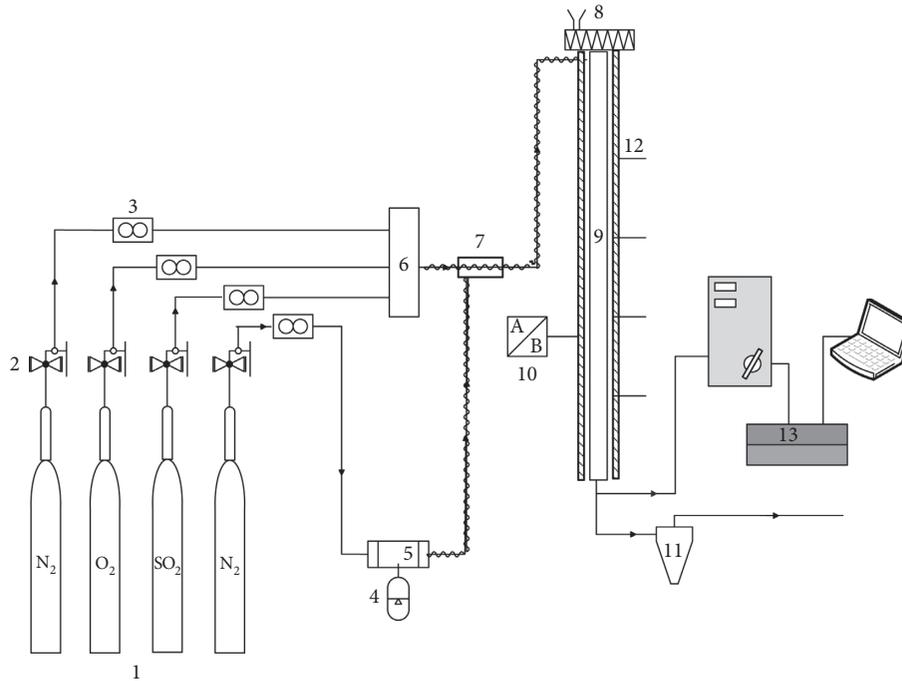


FIGURE 1: Diagram of the drop tube furnace. 1. Gas cylinder. 2. Pressure reducing valve. 3. Flowmeter. 4. Peristaltic pump. 5. Evaporator. 6 and 7. Mixer. 8. AC feeder. 9. Reactor. 10. Electric heater. 11. Cyclone. 12. Sampling hole. 13. Gas analyzer.

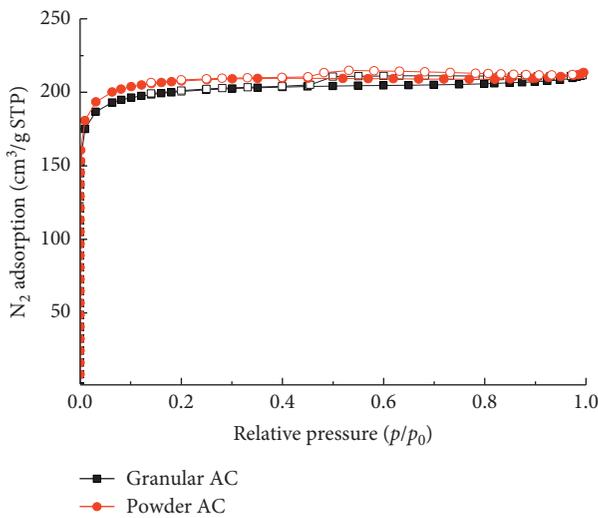


FIGURE 2: The N_2 adsorption isotherms of ACs.

TABLE 1: The pore structure parameters of ACs.

AC	S_{BET} (m^2/g)	S_{mic} (m^2/g)	V_{mic} (cm^3/g)	V_{mes} (cm^3/g)	L_{mic} (nm)
Powdered AC	624	541	0.29	0.023	0.699
Granular AC	605	514	0.27	0.037	0.696

3.2. Influence of AC/S Molar Ratio on SO_2 Adsorption. The influence of AC/S molar ratio on the adsorption of SO_2 by the powdered AC is shown in Figure 5. The flow rate of the simulated flue gas is 20 L/min, and therefore the gas

residence time in the reactor is about 6 seconds. In the first 1.2 seconds, the powdered AC quickly adsorbs SO_2 , and the amount of SO_2 adsorbed rapidly increases. And then the SO_2 adsorption rate by the powdered AC decreases and the amount of SO_2 adsorbed slowly increases. As shown in the reaction equations (1)–(5), the SO_2 in flue gas is first adsorbed on the surface of AC and then catalyzed to H_2SO_4 and H_2SO_4 is stored in pores of AC. The concentration difference between SO_2 in flue gas and SO_2 on the surface of the powdered AC is the driving force for the powdered AC to adsorb SO_2 . As the adsorption progresses, the active sites on the surface of the powdered AC are occupied by SO_2 , and the number of the vacant active sites decreases. The concentration of SO_2 on the surface of the powdered AC is increased while the concentration of SO_2 in flue gas is decreased. The concentration difference between SO_2 in flue gas and SO_2 on the surface of the powdered AC is therefore decreased and then the driving force for the powdered AC to adsorb SO_2 is also decreased. Therefore, the SO_2 removal rate by powdered AC increases slowly after 1.2 seconds.

Increasing the AC/S molar ratio increases the desulfurization efficiency of the powdered AC. When the AC/S molar ratio is increased from 100 to 345, the desulfurization rate by the powdered AC is increased from 28.11% to 70.24% at 1.2 seconds and is increased from 37.41% to 73.64% at 6.0 seconds, respectively. Increasing AC/S molar ratio indicates that the feed rate of the powdered AC is increased, the concentration of the powdered AC in flue gas is increased, more active sites are provided to adsorb SO_2 , and thereby the desulfurization efficiency is increased.

Increasing the AC/S molar ratio reduces the adsorption amount of SO_2 on the powdered AC. When the AC/S molar

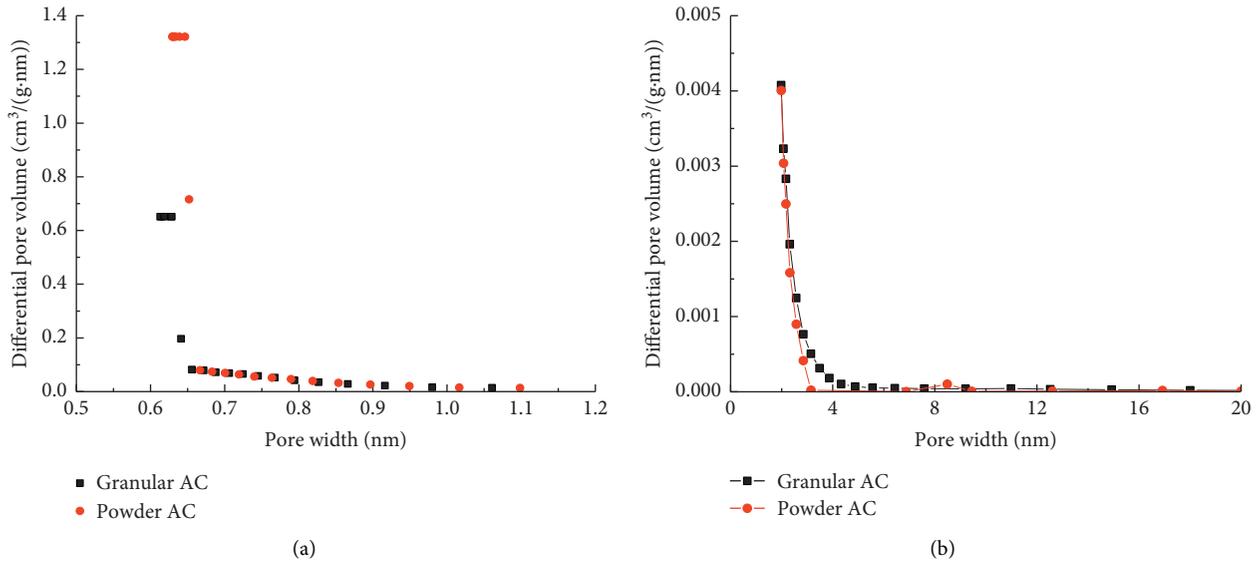


FIGURE 3: Pore-size distribution. (a) HK and (b) BJH.

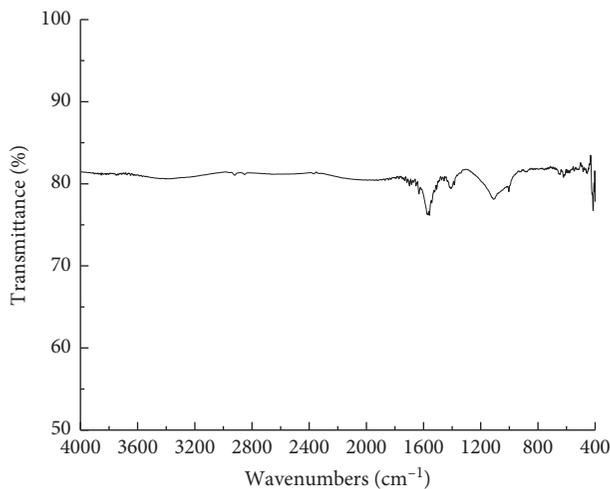


FIGURE 4: FT-IR spectra of the powdered AC.

ratio is increased from 100 to 345, the adsorption amount of SO₂ on the powdered AC is decreased from 12.87 mg/g to 9.23 mg/g at 1.2 seconds and is decreased from 17.13 mg/g to 10.11 mg/g at 6.0 seconds, respectively. When the AC/S molar ratio is 100, the concentration of the powdered AC in flue gas is lower and the concentration of SO₂ relative to the unit mass of powdered AC in flue gas is higher. The concentration difference between SO₂ in flue gas and SO₂ on the surface of the powdered AC is therefore increased and then the driving force for the powdered AC to adsorb SO₂ is also increased. When the AC/S molar ratio is 345, the concentration of the powdered AC in flue gas is higher and the competitive adsorption of SO₂ between the powdered ACs is increased, which reduces the adsorption quantity of SO₂ on the powdered AC. Zhang et al. have studied SO₂ removal by powdered AC in a CFB reactor and found that when the AC/S molar ratio increased, SO₂ removal efficiency increased

rapidly, but SO₂ adsorption quantity decreased [11]. The research results of this paper are consistent with those of Zhang.

3.3. Influence of the Reaction Temperature on SO₂ Adsorption.

The influence of the reaction temperature on the adsorption of SO₂ by powdered AC is shown in Figure 6. Increasing the reaction temperature reduces the desulfurization efficiency and the amount of SO₂ adsorbed by the powdered AC. When the reaction temperature is increased from 65°C to 95°C, the desulfurization rate by the powdered AC is decreased from 58.24% to 29.06% at 1.2 seconds and is decreased from 74.35% to 36.03% at 6.0 seconds, respectively. The amount of SO₂ adsorbed by the powdered AC is decreased from 10.74 mg/g to 5.36 mg/g at 1.2 seconds and is decreased from 13.71 mg/g to 6.64 mg/g at 6.0 seconds, respectively.

The AC/S molar ratio is 245 and SO₂, O₂, and H₂O concentration is 0.085%, 6%, and 8%.

As shown in reaction equations (1)–(5), the powdered AC is used as adsorbent and catalyst to convert SO₂ into H₂SO₄. The adsorption of SO₂ and the catalytic oxidation of SO₂ to H₂SO₄ by the powdered AC are all affected by reaction temperature. From the perspective of adsorption, the adsorption of SO₂ in the gas phase on the solid surface is an instantaneous process. With the reduction of the free energy of the system, it also loses its degree of freedom, resulting in a reduction in enthalpy. Regardless of physical adsorption or chemical adsorption, the adsorption process is always exothermic. Therefore, increasing temperature is not conducive to the adsorption of SO₂ by the powdered AC. From the perspective of surface catalytic oxidation reaction, there is the breakage and formation of chemical bonds, which requires certain temperature conditions. Therefore, increasing temperature is conducive to the catalytic oxidation

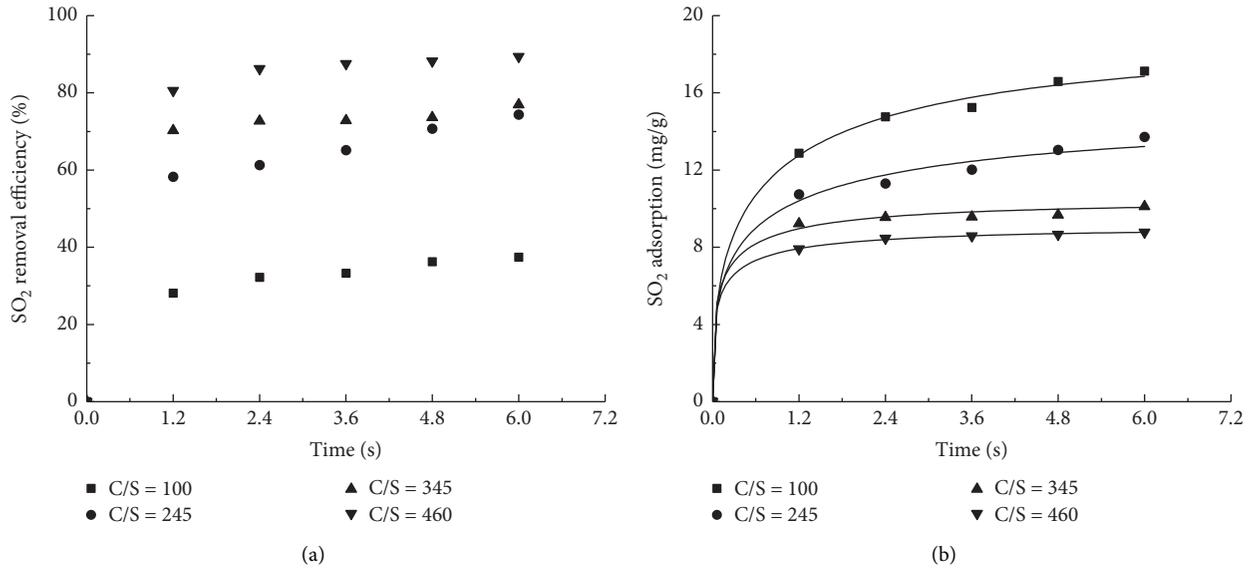


FIGURE 5: Influence of AC/S molar ratio on (a) SO₂ removal rate and (b) SO₂ adsorption. The temperature is 65°C and SO₂, O₂, and H₂O concentration is 0.085%, 6%, and 8%.

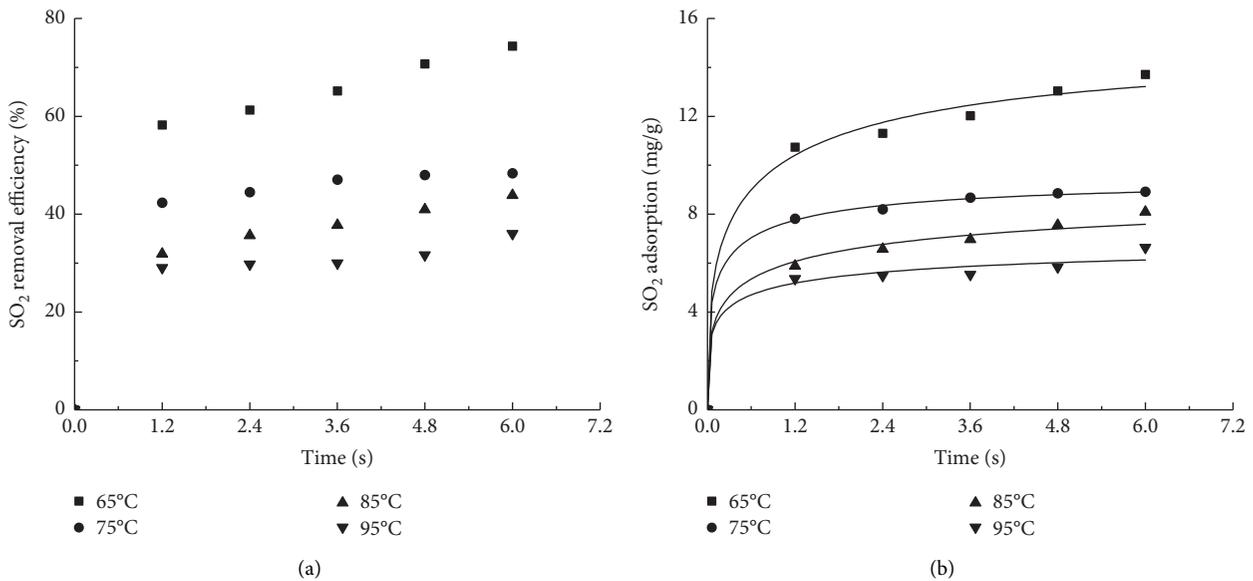


FIGURE 6: Influence of temperature on (a) SO₂ removal rate and (b) SO₂ adsorption.

reaction of SO₂ by the powdered AC. The temperature has a great influence on the physical adsorption of SO₂ by powdered AC. As the temperature increases, it reduces the physical adsorption of SO₂, which in turn affects the catalytic oxidation of SO₂ by the powdered AC. In addition, H₂O adsorption decreases with increasing temperature, and H₂SO₄ cannot be desorbed from the active sites in time [14].

3.4. Influence of SO₂ Concentration on SO₂ Adsorption. The influence of the concentration of SO₂ on the adsorption of SO₂ by powdered AC is shown in Figure 7. Increasing the concentration of SO₂ reduces the desulfurization efficiency

of the powdered AC. When the concentration of SO₂ is increased from 0.015% to 0.05%, the desulfurization rate of the powdered AC is decreased from 86.02% to 74.13% at 1.2 seconds and is decreased from 94.67% to 77.25% at 6.0 seconds, respectively. When the concentration of SO₂ is increased, a part of SO₂ could not be adsorbed by the powdered AC and therefore the desulfurization efficiency is decreased.

The temperature is 65°C, AC feeding rate is 2.634 g/min, and O₂ and H₂O concentration is 6% and 8%.

Increasing the concentration of SO₂ increases the amount of SO₂ adsorbed by the powdered AC. When the concentration of SO₂ is increased from 0.015% to 0.05%, the

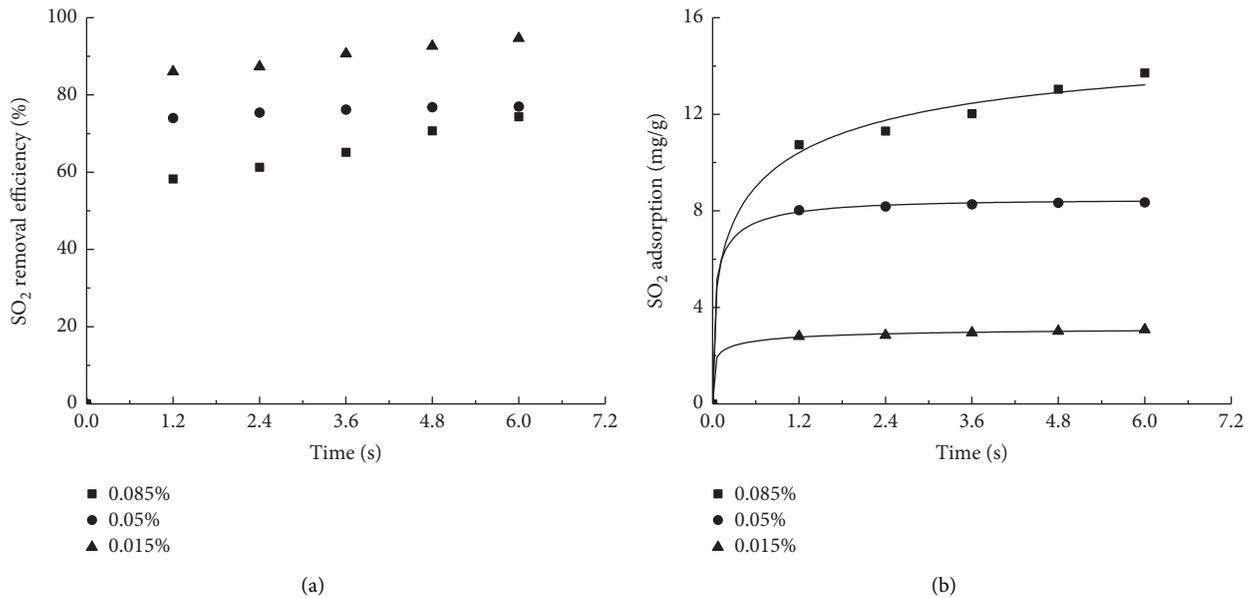


FIGURE 7: Influence of SO₂ concentration on (a) SO₂ removal rate and (b) SO₂ adsorption.

amount of SO₂ adsorbed by the powdered AC is increased from 2.80 mg/g to 8.03 mg/g at 1.2 seconds and is increased from 3.08 mg/g to 8.35 mg/g at 6.0 seconds, respectively. Increasing the concentration of SO₂ increases the concentration difference between SO₂ in flue gas and SO₂ on the surface of the powdered AC, and then the driving force for the powdered AC to adsorb SO₂ is also increased [15].

3.5. Influence of O₂ Concentration on SO₂ Adsorption. The influence of the concentration of O₂ on the adsorption of SO₂ by the powdered AC is shown in Figure 8. Increasing the concentration of O₂ increases the desulfurization efficiency and the amount of SO₂ adsorbed by the powdered AC. When the concentration of O₂ is increased from 0% to 6%, the desulfurization rate by the powdered AC is increased from 22.32% to 58.24% at 1.2 seconds and is increased from 25.76% to 74.35% at 6.0 seconds, respectively. The amount of SO₂ adsorbed by the powdered AC is increased from 4.06 mg/g to 10.74 mg/g at 1.2 seconds and is increased from 4.75 mg/g to 13.71 mg/g at 6.0 seconds, respectively.

There are two forms of adsorption of SO₂ on AC. One is weakly bonded SO₂, which is easy to be desorbed at low temperature, corresponding to physically adsorbed SO₂ and the other is strongly bonded SO₂ which can only be desorbed at high temperature, namely, SO₃, which is related to the active sites of catalytic oxidation on AC surface [16]. Pinero et al. have found that, in the absence of O₂, SO₂ was almost physical adsorption, and activated carbon materials could not oxidize SO₂ to SO₃ [16]. When the concentration of O₂ is 0%, SO₂ adsorbed on the surface of the powdered AC could not be oxidized to SO₃ and generated to H₂SO₄, which cannot empty the SO₂ adsorption center and hinder the adsorption of SO₂. At this time, SO₂ mainly exists in the form of physical adsorption, with lower adsorption amount and lower desulfurization efficiency. When the concentration of O₂ increases,

SO₂ adsorbed on the surface of the powdered AC is easily oxidized to SO₃, which interacts with H₂O to generate H₂SO₄, and H₂SO₄ could be desorbed from the active center regenerated to further adsorb SO₂, which increases the desulfurization efficiency and the amount of SO₂ adsorption.

3.6. Influence of H₂O Concentration on SO₂ Adsorption.

The influence of the concentration of H₂O on the adsorption of SO₂ by the powdered AC is shown in Figure 9. Increasing the concentration of H₂O increases the desulfurization efficiency and the amount of SO₂ adsorbed by the powdered AC. When the concentration of H₂O is increased from 0% to 8%, the desulfurization rate by the powdered AC is increased from 23.18% to 58.24% at 1.2 seconds and is increased from 26.94% to 74.35% at 6.0 seconds, respectively. The amount of SO₂ adsorbed by the powdered AC is increased from 4.27 mg/g to 10.74 mg/g at 1.2 seconds and is increased from 4.97 mg/g to 13.71 mg/g at 6.0 seconds, respectively.

Mochida et al. have studied the continuous adsorption of SO₂ by AC fiber at low temperature, and the product was aqueous H₂SO₄. Mochida has found that larger H₂O and O₂ concentrations in flue gas were conducive to the continuous removal of SO₂ in the form of aqueous H₂SO₄, and the rate control step was the aqueous H₂SO₄ desorption from the surface of AC fiber [2]. When the volume fraction of H₂O is 0%, SO₂ is oxidized to SO₃ by O₂ and SO₃ cannot be desorbed, which makes the active center invalid and cannot continue to adsorb SO₂. When the volume fraction of H₂O increases, SO₃ is easily combined with H₂O to form H₂SO₄ after SO₂ oxidized to SO₃ by O₂. Excess H₂O can elute H₂SO₄ from the active center. The eluted H₂SO₄ is stored in the micropores of the powdered AC, and the active center is regenerated to continue to adsorb and oxidize SO₂. The increase of the volume fraction of H₂O in flue gas has two main effects: on the one hand, the product SO₃ after SO₂

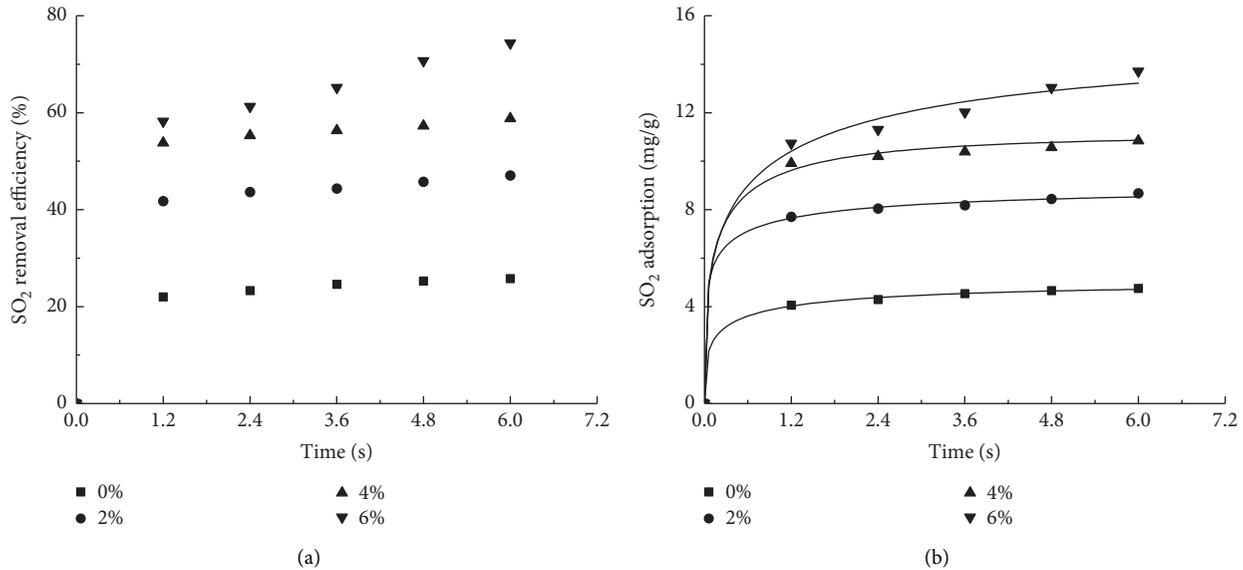


FIGURE 8: Influence of O₂ concentration on (a) SO₂ removal rate and (b) SO₂ adsorption. The temperature is 65°C, AC/S molar ratio is 245, and SO₂ and H₂O concentration is 0.085% and 8%.

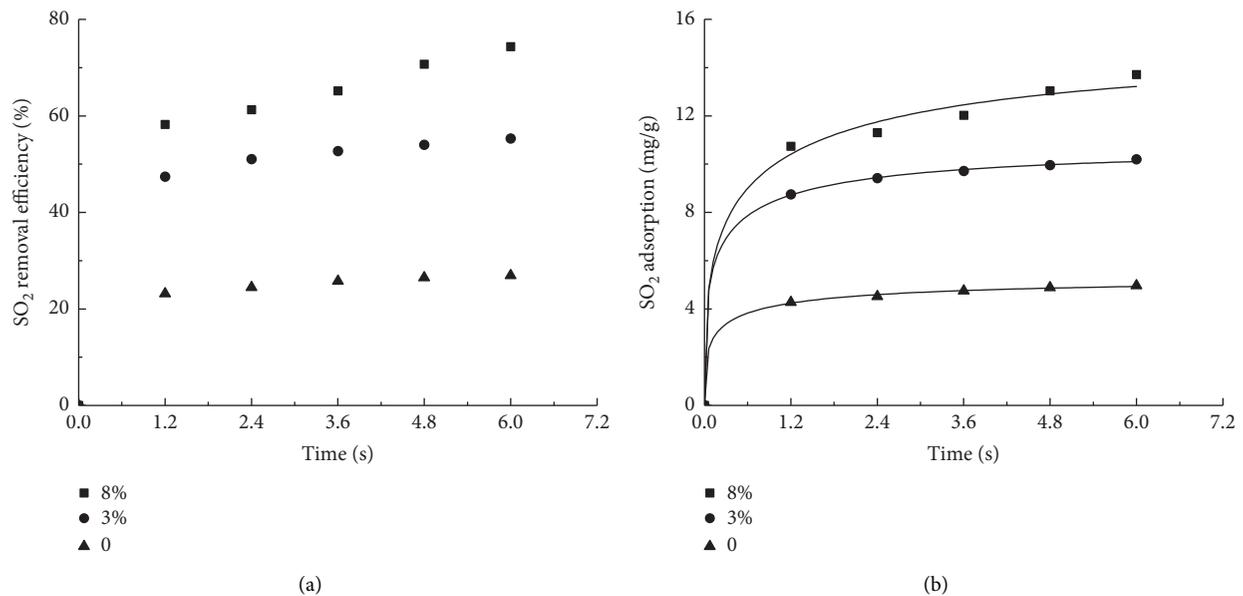


FIGURE 9: Influence of H₂O concentration on (a) SO₂ removal rate and (b) SO₂ adsorption. The temperature is 65°C, AC/S molar ratio is 245, and SO₂ and O₂ concentration is 0.085% and 6%.

oxidation is hydrated to H₂SO₄, and on the other hand, H₂SO₄ is eluted from the active center by excessive H₂O to regenerate active centers to facilitate the continuous adsorption of SO₂ [2, 7].

3.7. Influence of the Powdered AC Circulation Ratio on SO₂ Adsorption. The adsorption quantity of SO₂ is lower than the saturated adsorption capacity of SO₂ by the powdered AC under various working conditions [6, 15], so the recycling of the powdered AC plays a very important role in improving the desulfurization rate, reducing the amount of the powdered

AC, and improving the utilization rate of the powdered AC. In order to study the cyclic adsorption performance of the powdered AC, a material receiving device is arranged at the outlet of the reactor to collect the powdered AC, which is defined as the first adsorption. And then the collected powdered AC is sent to the reactor according to a certain AC/S molar ratio to measure the desulfurization performance, and the powdered AC is collected again at the material receiving device, which is defined as the second adsorption, in turn until the adsorption of SO₂ is saturated. The influence of cyclic adsorption of the powdered AC on the adsorption of SO₂ is shown in Figure 10. When the cyclic adsorption is

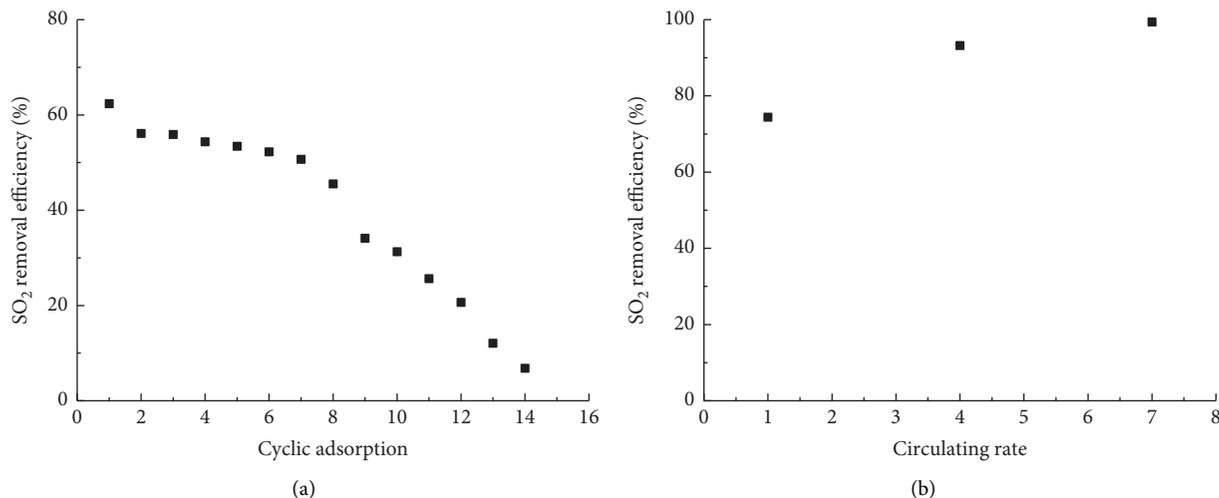


FIGURE 10: (a) Influence of cyclic adsorption on SO₂ removal rate. (b) Influence of circulating ratio on SO₂ removal rate. The temperature is 65°C, AC/S mole ratio is 300, and SO₂, O₂, and H₂O concentration is 0.1%, 6%, and 8%.

increased from the first to the eighth, the desulfurization rate by the powdered AC is decreased slowly from 62.35% to 45.53%; after that, the decreasing trend of the desulfurization rate is accelerated, and the desulfurization rate is decreased to 6.83% at the fourteenth.

With the increase of cyclic adsorption, SO₂ adsorption amount increases, and then H₂SO₄ generated gradually increases. Because H₂SO₄ is stored in AC pores, H₂SO₄ occupies the active sites of AC, and the desulfurization rate decreases with cyclic adsorption increasing until the adsorption of SO₂ is saturated.

The circulation ratio refers to the ratio of the amount of material captured by the material separator and returned to the reactor to the amount of material supplied. In this paper, the influence of the circulation of the powdered AC on the desulfurization efficiency is simulated under the condition that the molar ratio of fresh powdered AC to SO₂ in flue gas is certain. The results are shown in Figure 10(b). Increasing the circulation ratio of the powdered AC increases the desulfurization efficiency. The desulfurization efficiency of the powdered AC is 62.35% when the powdered AC is not recycled. The desulfurization efficiency is increased to 99.42% when the circulation ratio of the powdered AC is 7. When the AC/S mole ratio is 300 and the powdered AC circulating ratio is 7, SO₂ can be removed effectively.

Increasing the circulation ratio of the powdered AC increases the concentration of AC in reactor and the number of active sites, which is conducive to the removal of SO₂.

3.8. The SO₂ Adsorption Kinetics. The Bangham model can be used to fit the adsorption process where multiple adsorption mechanisms exist, such as gas film diffusion, surface reaction, and intraparticle diffusion. This model was obtained by time compensated pseudo-first-order kinetics model, which made it have good adaptability in various adsorption processes. The Bangham model is shown in the following equation [17–19]:

TABLE 2: The parameters of Bangham model of Figure 5(b).

AC/S molar ratio	100	245	345	460
q_e (mg/g)	18.7	14.9	10.2	9.0
K	1.06	1.13	1.88	2.01
N	0.44	0.38	0.37	0.35
R^2	0.998	0.993	0.997	0.999

TABLE 3: The parameters of Bangham model of Figure 6(b).

Temperature (°C)	65	75	85	95
q_e (mg/g)	14.9	9.3	8.9	6.9
K	1.13	1.68	1.08	1.33
N	0.38	0.35	0.32	0.29
R^2	0.993	0.999	0.991	0.980

TABLE 4: The parameters of Bangham model of Figure 7(b).

SO ₂ concentration (%)	0.085	0.05	0.015
q_e (mg/g)	14.9	8.5	3.2
K	1.13	2.65	1.97
N	0.38	0.37	0.26
R^2	0.993	0.999	0.999

$$q_t = q_e(1 - e^{-kt^n}), \quad (8)$$

where q_t is the adsorption amount of SO₂ on the powdered AC at time t , q_e is the saturation adsorption capacity of SO₂ on the powdered AC, mg/g, and k and n are constants.

Figures 5(b), 6(b), 7(b), 8(b), and 9(b) show the fitting results of the Bangham model. Tables 2–6 show the parameters of the Bangham model. The calculated values of the Bangham adsorption kinetics model agree well with the experimental values, and the fitting correlation coefficient R^2 is higher, all of which are above 0.98. This indicates that SO₂

TABLE 5: The parameters of Bangham model of Figure 8(b).

O ₂ concentration (%)	0	2	4	6
q_e (mg/g)	5.0	8.9	11.1	14.9
K	1.54	1.84	1.88	1.13
N	0.35	0.29	0.41	0.38
R^2	0.999	0.999	0.998	0.993

TABLE 6: The parameters of Bangham model of Figure 9(b).

H ₂ O concentration (%)	0	3	8
q_e (mg/g)	5.2	10.6	14.9
K	1.58	1.61	1.13
N	0.34	0.35	0.38
R^2	0.993	0.999	0.993

adsorption on the powdered AC in the drop tube furnace is controlled by multiple mechanisms, such as gas film diffusion, intragranule diffusion, and surface reaction. The SO₂ adsorption kinetics can be predicted by Bangham model.

4. Conclusions

The SO₂ adsorption kinetics on powdered AC was performed in a laboratory-scale drop tube furnace. The results show that the powdered AC rapidly adsorbs SO₂ in the initial 1.2 seconds and then the amount of SO₂ adsorption slowly increases. The SO₂ adsorption kinetics can be predicted by Bangham model.

The SO₂ removal rate increases with the increasing of AC/S molar ratio, the decreasing of adsorption temperature, and inlet SO₂ concentration. The adsorption amount of SO₂ by the powdered AC increases with the increasing of the inlet SO₂ concentration, the decreasing of AC molar ratio, and adsorption temperature. The O₂ and H₂O are beneficial to SO₂ removal by the powdered AC.

The recycling of the powdered AC plays a very important role in improving the desulfurization rate, reducing the amount of the powdered AC and improving the utilization rate of the powdered AC. SO₂ removal rate drops to 6.83% after 14 cyclic adsorptions. The powdered AC circulation increases SO₂ removal rate from 62.35% to 99.42% with AC circulation ratio = 7.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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