Removal of Chlorine Ions from Desulfurization Wastewater by Modified Fly Ash Hydrotalcite

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ABSTRACT: The effective removal of chlorine ion from the desulfurization slurry is of great significance to the stable operation of the desulfurization system. Modified fly ash hydrotalcites were prepared by alkali/acid-combined roasting and microwaving and used as an adsorbent for chlorine ion in desulfurized wastewater. The specific surface area and porosity of different adsorbents were analyzed by X-ray diffraction (XRD) and scanning electron microscopy (SEM). The impacts of pH, temperature, adsorbent dosage, and adsorption shaking time on adsorption performance were investigated. Results showed the alkali-combined roasting-modified fly ash hydrotalcite has the optimum removal effect on Cl\(^-\). The optimal adsorption performance was achieved when the pH was 8, the adsorption temperature was 60 \(^\circ\)C, the mass concentration of adsorbent was 10 g/L, the adsorption shaking time was 180 min, and the removal percentage of Cl\(^-\) was 68.1%. The adsorption isotherm was consistent with the Langmuir isotherm model, and the adsorption saturation was 694.4 mg/g, which belonged to monolayer adsorption.

1. INTRODUCTION

Desulfurization wastewater contains a considerable amount of suspended solids (e.g., gypsum particles, SiO\(_2\), aluminum, and iron hydroxide), inorganic salts, and trace heavy metals, such as arsenic, cadmium, chromium, and mercury, thus seriously harming the environment if directly discharged. The dissolution of chlorides in the flue gas will increase the chloride ion concentration in the desulfurization absorbent. This increase will decrease the desulfurization rate and affect the gypsum quality, resulting in the corrosion of desulfurization systems. After the triple box process of "neutralization, coagulation, and precipitation", the content of heavy metals and suspended solids in desulfurization wastewater is reduced but that of chlorine remains high. The 2017 guidelines on feasible technologies for the prevention and control of pollution in thermal power plants (HJ2301-2017) indicated that the key to achieving a near-zero discharge of wastewater is to realize a zero discharge of desulfurization wastewater. The main methods currently used to remove Cl\(^-\) from wastewater include electric coagulation, ion exchange, and adsorption. Adsorption is regarded as one of the effective ways to uptake Cl\(^-\) from wastewater due to its advantages of simple operation, recyclability, etc.

Fly ash is a byproduct of coal-fired power plants. China is a large producer and user of coal. The coal-fired power generation of China can produce hundreds of millions of tons of fly ash yearly. Fly ash is characterized by its low cost, easy acquisition, porous physical tissue, large specific surface area, and high adsorption activity. Therefore, the treatment of harmful substances in wastewater using fly ash has been widely investigated. Studies on the removal of phosphorus from wastewater by fly ash began in 1980 when Kuziemskzka applied the water extract of brown fly ash as a coagulant to treat phosphorus in wastewater. Mohan used fly ash as a cheap adsorbent to treat heavy metal ions in urban solid waste leachates. Studies have shown that modified fly ash has a larger specific surface area and a stronger adsorption capacity than those of unmodified fly ash. Therefore, using modified fly ash to treat some harmful substances is possible due to its strong adsorption performance. Wang used the H\(_2\)SO\(_4\) impregnation method to modify raw coal fly ash (RCFA) and used the modified RCFA (ACFA) as a Fenton-like catalyst to treat polymer flooding wastewater (PW) under microwave prestrengthening conditions. The fly ash was modified by nitric acid at room temperature, and the adsorption performance of the modified fly ash to methylene blue was significantly improved. Nguyen used 2-mercapto-benzothiazole (MBT) and sodium dodosulfate (SDS) as surfactants and treated fly ash with sodium hydroxide to investigate the morphology, composition, and structural changes of modified and unmodified fly ash as well as the adsorption behavior of Cd\(_{2+}\) and Hg\(_{2+}\). Sahoo studied the removal of heavy metals from acidic mine wastewater by alkali-modified fly ash. Research by Li and others found that the acid-
modified fly ash method has the best effect, with the chemical oxygen demand (COD) removal rate reaching more than 90% and the wastewater after degradation meeting the discharge standard. Malarvizhi used 4 mol/L NaOH and fly ash that were mixed and stirred at 90 °C for 1 h, washed with deionized water, and then dried. The adsorption of heavy metals in wastewater indicates that alkali can cause the dissolution and precipitation of fly ash. Specific surface area and activity of fly ash increased due to the dissolution of fly ash acidic oxides, and the adsorption performance of heavy metals in wastewater is enhanced. Fly ash, as a water treatment chemical, is mostly used to remove impurities in water, heavy metals, or decolorization experiments. Two or more modification methods are often combined to modify fly ash and improve its adsorption performance. The fly ash modified by microwave combined with alkali had excellent adsorption properties for Hg^{2+} and Cr^{6+} in aqueous solutions. Shi investigated the sorption of chlorinated hydrocarbons from synthetic and natural groundwater by organohydrotalcites. However, studies on using fly ash as a Cl^{-} adsorbent are currently limited.

In this paper, modified fly ash hydrotalcites were prepared by alkali/acid-combined roasting and microwaving and used as adsorbents for chlorine ion in desulfurized wastewater. The optimal preparation conditions and adsorption isotherms of adsorbents were studied, and the influence of various preparation conditions on the removal rate was analyzed.

2. RESULTS AND DISCUSSION

2.1. Determination of the Optimum Modification Method. 2.1.1. Adsorbent Characterization. Figure 1 shows the surface morphology of the original fly ash hydrotalcite and the scanning electron microscopy (SEM) diagram of the fly ash hydrotalcite modified by four modification methods. Figure 1a shows raw ash, and Figure 1b–e is the SEM diagrams of alkali-combined roasting, acid-combined roasting, alkali-combined microwave, and acid-combined microwave modification. Figure 1 shows that the surface of modified fly ash hydrotalcite is rougher than that of the original fly ash. The porous structure is also more obvious, greatly changing its original surface morphology, and the specific surface area and pore volume are greatly improved. In addition, the SEM diagrams corresponding to different modification methods are different. The surface of the particle is smoother, and picture b is the roughest, followed by c, d, and e. The specific surface area from large to small is b > c > d > e > a (see Table 1). Therefore, the

| Table 1. Specific Surface Area and Pore Volume Analysis |
|--------------------------------------------------------|
| species | specific surface area (m²/g) | pore volume (cc/g) |
| the original gray | 5.310 | 0.007 |
| alkali + roasting modification | 24.579 | 0.075 |
| acid + roasting modification | 19.899 | 0.033 |
| alkali + microwave modification | 14.155 | 0.040 |
| acid + microwave modification | 11.767 | 0.011 |
adsorption capacity is also different. The alkali-combined roasting modification method has a more evident removal percentage of Cl\(^{-}\) compared with that of other modification methods. Thus, the SEM surface of this method is rough and its porous structure is evident. Therefore, the adsorption effect of this method is better.

Table 1 shows the specific surface area and pore volume of fly ash and modified fly ash hydrotalcite. The specific surface area of the adsorbent modified by alkali roasting was 24.579 m\(^2\)/g, which was considerably larger than that of the original fly ash and increased by approximately fivefold. The OH\(^{-}\) dissolved from fly ash and Na\(^{+}\) and K\(^{+}\) in alkaline modifier destroyed Si–O and Al–O bonds in the fly ash vitreous body, depolymerized the silicate glass network, promoted the formation of zeolite skeleton from fly ash, and increased many new active points in alkali modification, as shown in Figure 2; thus, the physical and chemical adsorption performances of fly ash are improved.\(^{28,29}\)

Figure 2. Aircraft layout of alkali-modified fly ash.

The volume and the surface area of fly ash increased during high-temperature roasting and modification. This increase could be attributed to the melting of fusible substances in fly ash, the destruction of the glass network structure, the depolymerization of the original high polymer silicate network, and the loose and porous fly ash particles. Simultaneously, the water on the surface of fly ash particles at the beginning of high-temperature roasting modification was evaporated as temperature increased; this phenomenon exposed additional adsorption activity points on the fly ash surface and promoted the improvement of the adsorption capacity of fly ash.\(^{30}\)

2.1.2. Removal Percentage of Cl\(^{-}\) by Four Adsorbents. Figure 3 shows that the removal percentages of Cl\(^{-}\) using the four modified methods were 56.9, 45.2, 39.7, and 19.2%. These results showed that adsorbent A had the optimum removal effect on Cl\(^{-}\) in the desulfurization wastewater. Strong alkali can destroy the Si–O–Si and Si–O–Al network structure in depolymerized fly ash and can act as an activator to increase the activity of fly ash and improve the physical and chemical adsorption properties of fly ash. The high-temperature roasting makes the moisture inside the fly ash evaporate, and more pores are formed on the fly ash surface, which increases the surface active site of the fly ash. Therefore, the alkali-combined roasting-modified fly ash hydrotalcite was used in the subsequent optimization experiments of adsorption conditions.

2.2. Optimization of Adsorption Conditions. 2.2.1. Influence of pH. The wastewater with an initial mass concentration of 10 g/L Cl\(^{-}\) was prepared, and the pH of the wastewater was adjusted to 5, 6, 7, 8, 9, and 10. A total of 1 g of adsorbent was added to 100 mL of desulfurization wastewater, which was shaken for 2 h at a constant temperature of 40 °C in the water bath and then filtered. Figure 4 shows the removal percentage of Cl\(^{-}\) at different pH values.

Figure 3. Cl\(^{-}\) removal percentages by the four modification adsorbents.

The points of zero charge (pHPZC) of fly ash, hydrotalcite, and the alkali-combined roasting-modified fly ash hydrotalcite are 4.86, 11.21, and 8.52, respectively.\(^{31,32}\) As can be seen from Figure 4, initially, the removal percentage of Cl\(^{-}\) increased significantly with pH. The removal percentage reached the maximum at pH 8 and then decreased with the increase in pH. Because fly ash hydrotalcite is alkaline. In an acidic environment, fly ash can react chemically and destroy the porous structure. The pH value of modified fly ash hydrotalcite adsorbent cannot be less than 4 because its interlayer structure is destroyed and its adsorption performance is reduced. In a strongly alkaline environment, the OH\(^{-}\) in the solution combines with part of the adsorption point when the pH is considerably high, and competitive adsorption occurs with Cl\(^{-}\), reducing the dechlorination rate.

2.2.2. Influence of Temperature. Figure 5 shows that 1 g of adsorbent was placed in 100 mL of desulfurization wastewater with an initial mass concentration of 10 g/L of Cl\(^{-}\), and the pH of the waste solution was adjusted to 8. The solution was
shaken in a water bath at 20, 30, 40, 50, 60, 70, and 80 °C for 2 h and then filtered. The results show that the removal percentage of Cl\(^-\) increased first and then decreased as the oscillation temperature increased. The removal percentage reached the maximum value at 60 °C. This result shows that the removal percentage of ions increases with the temperature. This finding may be attributed to the physical adsorption at low temperatures and the temperature increase for easy desorption, thereby decreasing the removal percentage. The chemical adsorption, adsorption site activity, and removal percentage will all increase at 30–60 °C. The molecular thermal movement will be violent when the oscillation temperature continues to rise. This phenomenon results in the easy desorption of adsorbed particles, which are not conducive to adsorption and decreases the removal percentage.

2.2.3. Influence of Input and Dosage. Modified fly ash hydrotalcite at 0, 0.2, 0.4, 0.6, 0.8, 1, 1.2, and 1.4 g was added to 100 mL of wastewater containing the initial mass concentration of 10 g/L Cl\(^-\) at pH 8. The water bath was maintained at a constant temperature of 60 °C for 2 h and then filtered. Figure 6 shows the effect of different amounts of fly ash on Cl\(^-\) removal percentages in desulfurization wastewater.

**Figure 6. Effect of adsorbent dosage on the adsorption rate.**

Figure 6 shows that with the increase in modified fly ash additive quantity, the Cl\(^-\) removal percentage continues to rise with the additional mass concentration of 10 g/L modified fly ash. The removal percentage no longer increased when the adsorbent additive mass concentration continued to increase to 14 g/L. This phenomenon may be observed when the Cl\(^-\) concentration in the water for the reduction of the concentration gradient is no longer evident and the diffusion flux is close to 0. Thus, modified fly ash adsorbent adsorption reaches an equilibrium of Cl\(^-\).

2.2.4. Influence of Shaking Time. A total of 1 g of adsorbent was added to 100 mL of desulfurization wastewater with 10 g/L initial mass concentration of Cl\(^-\) at pH 8, and the shaking time was set as 60, 120, 180, 240, and 300 min at an oscillation temperature of 60 °C. Figure 7 shows the effect of different shaking times on the Cl\(^-\) removal percentages in desulfurization wastewater.

**Figure 7. Effect of shaking time on adsorption rate.**

Figure 7 shows that the general trend of the Cl\(^-\) removal percentage increased after the first reduction as the adsorption time increased and reached the maximum at 180 min. The removal percentage slightly dropped after 180 min. The results show that increasing the stirring time can raise the contact time and improve the adsorption effect. However, the adsorption amount reached saturation due to a long time. At this temperature, increasing the stirring time will not increase the adsorption rate. Thus, the removal percentage is reduced. Prolonged adsorption time is not conducive to the removal of Cl\(^-\). Thus, the optimum shaking time is 180 min.

Therefore, the optimum preparation conditions are as follows: modification method using alkali-combined roasting (the ratio of fly ash to hydrotalcite is 2:1), mass concentration of fly ash of 10 g/L, constant-temperature shaking time of water bath of 180 min, initial mass concentration of Cl\(^-\) of 10 g/L, oscillation temperature of 60 °C, waste liquid pH of 8, and adsorption rate of up to 68.1%.

2.3. Adsorption Isotherm. Adsorption isotherm refers to a curve that reflects the relationship of the amount of solute \(Q_e\) adsorbed on the unit mass by adsorbents at constant temperatures in a certain volume of solution and the corresponding equilibrium concentration of the solution \(C_e\). Adsorption isotherms can be used to describe the adsorption characteristics of the adsorbents. The Freundlich and Langmuir adsorption isotherms are two widely used adsorption isotherms in environmental chemistry.

(1) Freundlich empirical formula

\[
Q_e = \frac{x}{m} = Kc_e^{1/n}
\]

where \(Q_e\) is the adsorption capacity of the adsorbent, mg/g; \(C_e\) is the equilibrium concentration, mg/L; 1/n is the nonlinear coefficient (1/n is a constant describing the adsorption
strength); $0.1 < 1/n < 0.5$ is easily adsorbed, $1/n > 2$ is difficult to adsorb; and $k$ is the Freundlich constant.

The above equation is usually expressed as a logarithmic equation of a line.

$$\ln Q_e = \ln K + \frac{1}{n} \ln C_e$$  \hspace{1cm} (2)

(2) Langmuir empirical formula

$$\frac{1}{Q_e} = \frac{1}{X_m b} \times \frac{1}{C_e} + \frac{1}{X_m}$$  \hspace{1cm} (3)

where $Q_e$ is the adsorption capacity of the adsorbent, mg/g; $C_e$ is the equilibrium concentration, mg/L; $X_m$ is the saturated adsorption of adsorbent, mg/g; and $b$ is the constant related to adsorption energy. Figure 8 shows the adsorption isotherms of modified fly ash and hydrotalcite at 25 °C. Figures 9 and 10 show the Freundlich and Langmuir adsorption isotherms, respectively. The Langmuir model represents monolayer adsorption at specific homogeneous sites, while the Freundlich equation applies to multilayer adsorption. Moreover, the adsorption characteristics of modified fly ash hydrotalcite are more consistent with the Langmuir isotherm model, indicating that the adsorption of Cl\textsuperscript{−} by modified fly ash belongs to monolayer adsorption. The adsorption saturation is 694.4 mg/g. Table 2 shows the Brunauer–Emmett–Teller (BET) surface areas and the adsorption capacity of different adsorbents.

| adsorbents                          | specific surface area (m\textsuperscript{2}/g) | adsorption capacity (mg/g) |
|------------------------------------|-----------------------------------------------|---------------------------|
| alkali + roasting-modified fly ash hydrotalcite | 24.58                                         | 694.4                     |
| OBC1000-3-10                       | 62.26                                         | 4.34                      |
| OBC1000-3-15                       | 86.52                                         | 6.76                      |
| KOBC15                             | 303.59                                        | 11.13                     |

3. CONCLUSIONS

The increase in chloride ion concentration in the desulfurization absorbent will decrease the desulfurization efficiency and affect the gypsum quality. The effective removal of chlorine ion from the desulfurization slurry is of great significance for the stable operation of the desulfurization system. Modified fly ash hydrotalcites were prepared by alkali/acid-combined roasting and microwaving and used as adsorbents of chlorine ion in the desulfurized wastewater. The optimum preparation conditions for modifying fly ash hydrotalcite absorbent are as follows: joint alkali-roasting modification method (fly ash and hydrotalcite ratio is 2:1), adsorbent concentration of 10 g/L, water bath temperature shaking time of 180 min, Cl\textsuperscript{−} initial mass concentration of 10 g/L, oscillation temperature of 60 °C, liquid optimum pH of 8, and adsorption rate reaching 68.1%.

Fly ash hydrotalcite absorbent modified using joint alkali roasting demonstrated the following: a large surface area and pore volume, a strong adsorption capability, consistent Cl\textsuperscript{−} adsorption isotherm with the Freundlich and Langmuir

![Figure 8. Adsorption isotherm of Cl\textsuperscript{−} by the modified adsorbent.](image)

![Figure 9. Freundlich isotherm model.](image)

![Figure 10. Langmuir isotherm model.](image)
adsorption isotherms, and correlation coefficient $R^2$ of 0.9826 and 0.9931.

4. MATERIALS AND METHODS

4.1. Preparation of Modified Fly Ash Hydrotalcite. The fly ash used in the experiment was obtained from the Baoding power plant. The chemical composition of the samples was determined using the test method for the analysis of the coal ash according to the Chinese standard GB/T 1574-2007, as shown in Table 3.

Table 3. Chemical Composition of Fly Ash

| chemical composition | content (%) |
|----------------------|-------------|
| SiO$_2$              | 54.1        |
| Fe$_2$O$_3$          | 17.5        |
| Al$_2$O$_3$          | 11.6        |
| CaO                  | 11.3        |
| TiO$_2$              | 2.69        |
| K$_2$O               | 1.18        |
| MgO                  | 0.192       |
| BaO                  | 0.162       |
| MnO                  | 0.126       |
| others               | 1.15        |

Table 3 shows that the contents of SiO$_2$, Fe$_2$O$_3$, and Al$_2$O$_3$ in fly ash account for 80%. Fly ash contains a large number of irregular and closed pores rich in active vitreous particles, such as SiO$_2$ and Al$_2$O$_3$. These vitreous particles contain many small active channels, thus providing a large specific surface area and good activity and increasing their adsorption capacity. The typical molecular composition of talc in the experiment is Mg$_6$Al$_2$(OH)$_{16}$CO$_3$.4H$_2$O, also known as layered bimetallic hydroxide, which is a kind of anion clay with a layered structure similar to the regular octahedral structure of brucite Mg(OH)$_2$.

Fly ash and hydrotalcite are taken as raw materials in the present study. After mixing fly ash and hydrotalcite in the 2:1 ratio, four modification methods are adopted to produce four required adsorbents to determine the optimal scheme, as shown in Table 4.

4.2. Feature Analysis. The wastewater used in the experiment was simulated wastewater, which was dissolved in 1 L of deionized water with 10 g of NaCl, and the mass concentration of Cl$^-$ was 10 g/L. The main instruments used in this experiment are listed in Table 5. The specific surface area and pore volume of the adsorbent were tested by the porous physical adsorption instrument of the American Quantachrome Electronics in Japan. The BET (pore size) is defined as the BET surface area and pore volume of the adsorbent were tested by the porous physical adsorption instrument of the American Quantachrome Electronics in Japan.

NaCl standard solution of 10 mol/L was used to prepare a Cl$^-$ standard solution with a concentration of 6.0 mol/L. The Cl$^-$ concentration was determined using the ion-selective electrode method. The correlation coefficient $R^2$ is 0.999 based on the illustrated standard curve. The line equation in the standard curve illustration is obtained as follows: $y = 0.949 + 8.9157x$. The linear regression equation is $y = -53.835x + 89.157$. The correlation coefficient $R^2 = 0.9999$.

4.3. Adsorption Experiment. A total of 100 mL of simulated waste liquid was added to 250 mL of the tapered flask. A total of 1 g of prepared adsorbent was also added, and the nanotube was placed onto a speed-regulating multi-purpose oscillator to vibrate for 60 min and then stopped. The simulated waste liquid was allowed to stand for a period and then filtered. The clear liquid was taken to determine the content of Cl$^-$. The concentration of Cl$^-$ was identified through the ion-selective electrode method. The SEM images were obtained by field emission scanning electron microscopy and manufactured by JEOL Electronics in Japan.

Table 4. Preparation Schemes of Four Adsorbents

| adsorbent | modification methods | specific steps |
|-----------|---------------------|---------------|
| A         | alkali-combined roasting modification | Fly ash hydrotalcite was placed into 10 wt % sodium hydroxide solution to shake for 1 h and then filtered and roasted in a muffle furnace at 450 °C for 2 h to obtain adsorbent A. |
| B         | alkali combined with microwave modification | Fly ash hydrotalcite was placed into 10 wt % sodium hydroxide solution to shake for 1 h and then filtered. Microwave irradiation was conducted at 600 W, 60 °C for 10 min and then maintained at room temperature for 2 h. Adsorbent B was obtained after cooling to room temperature. |
| C         | acid-combined roasting modification | Fly ash hydrotalcite was placed into 10 wt % sulfuric acid solution to shake for 1 h and then filtered. Microwave irradiation was conducted at 600 W, 60 °C for 10 min and then maintained for 2 h at room temperature. Adsorbent D was obtained after cooling to room temperature. |
| D         | acid combined with microwave modification | Fly ash hydrotalcite was placed into 10 wt % sulfuric acid solution to shake for 1 h and then filtered. Microwave irradiation was conducted at 600 W, 60 °C for 10 min and then maintained for 2 h at room temperature. Adsorbent D was obtained after cooling to room temperature. |

Table 5. Main Experimental Instruments

| the instrument | model | manufacturer |
|----------------|-------|--------------|
| microwave synthesis reaction apparatus | XH-MC-1 | Beijing Xianghu Technology Development Co., Ltd. |
| muffle furnace | SA2-9-17TP | Nanyang Xinyu Furnace Industry Co., Ltd. |
| vacuum drying oven | ZDF6050 | Shanghai Qian Scientific Instrument Co., Ltd. |
| speed-regulating multipurpose oscillator | HY-4 | Changzhou Runhua Electric Appliance Co., Ltd. |
| ion meter | PXS-270 | Shanghai Electronic Science Instrument Co., Ltd. |
| electronic balance | FA2004B | Shanghai Youke Instrument Co., Ltd. |
| thermostatic magnetic stirrer | HJ-6AB | Changzhou Runhua Electric Appliance Co., Ltd. |
| BET (porous physical adsorption instrument) | QUANTACHROME | Conta EVO |
| SEM (field emission scanning electron microscope) | JSM-7800F Prime | Japanese Electronic JEOL |
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