Preparation of Honeycomb-Structured Activated Carbon–Zeolite Composites from Modified Fly Ash and the Adsorptive Removal of Pb(II)

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ABSTRACT: In this paper, fly ash (FA) was successfully prepared into a honeycomb carbon–zeolite composite (CZC) with good adsorption and used for the removal of Pb(II) by a two-step method. Compared with general FA, the honeycomb structure of the CZC resulted in a ∼6× increase in the specific surface area, and the average pore size increased from 3.4 to 12.7 μm. The maximum adsorption capacity of CZCs for Pb(II) reached 185.68 mg/g in 40 min. The experimental data for the adsorption of Pb(II) by CZC showed that the results were in good agreement with the Langmuir adsorption model. The adsorbent prepared in this study has good application prospects in wastewater treatment and provides a new method for the resource recovery of FA.

INTRODUCTION

Heavy metals are metal elements with a specific gravity greater than five (occasionally greater than four). They are widely present in the natural environment in many forms and a wide range of about 45–60 species, such as copper, zinc, chromium, mercury, lead, cadmium, arsenic, etc.1,2 With the rapid development of electroplating, oil refining, paint production, battery manufacturing, and other industries, heavy metal ions accumulate and migrate in the environment, such as water bodies, atmosphere, and soil, seriously polluting the environment and posing a greater risk to the ecosystem and human health than can be ignored. Heavy metals in water bodies enter the human body through the step-by-step transfer and enrichment of the food chain, and low concentrations of heavy metals can increase millions of times through bioenrichment. Human health is seriously threatened by chronic and acute poisoning, carcinogenesis, teratogenesis, and metamorphosis.3,4

For example, cadmium accumulation in the human body can be harmful to the kidneys, liver, and bones and cause people to suffer from diabetes, hypertension, bone pain, etc. In serious cases, a life-threatening and painful disease has been caused by world-famous heavy metal cadmium poisoning events.5,6 Mercury can damage the human nervous system. The damage is irreversible and can lead to cerebellar dysmotility, dysphonia, reduced visual field, and in severe cases mental confusion, spams, and even death.7 Although copper is an essential trace element, acute copper toxicity can lead to intestinal toxicity, while chronic copper toxicity can damage the liver, kidneys, and central nervous system and can lead to brain tissue lesions in severe cases. Lead can harm the human nervous system, the bone marrow hematopoietic system, the immune system, kidneys, and the reproductive system. Additionally, dizziness, memory loss, insomnia, and other symptoms will appear when the level of lead in the blood reaches 60–80 μg per 100 mL. Lead can also affect the liver, leading to hepatomegaly and jaundice, and in serious cases it can lead to cirrhosis and liver necrosis. Autonomic dysfunction, lead nephropathy, and impaired reproductive function are also some of the hazards that lead can pose to human body. Lead can also cause harm to the human body.

Most industrial wastewater contains high concentrations of Pb(II); moreover, Pb(II) is non-biodegradable and persists in the ecosystem.8 The Chinese Environmental Protection Administration (EPA) stipulates that the Pb concentration in water sources should not exceed 0.05 mg/L. Therefore, there is an urgent need to develop Pb(II) removal processes in wastewater treatment.

Several methods are available to mitigate heavy metal contamination in aqueous solutions, including electrolysis,
oxidation—reduction, and membrane separation.\textsuperscript{9,10,11} However, many of these methods have high operating costs and low separation efficiencies in wastewater with a low lead content. Therefore, adsorbent separation processes have attracted the attention of researchers because of their relatively simple operational processes, low potential for byproduct formation, low cost, and easy regeneration procedures.\textsuperscript{12,13} Many adsorbents have been synthesized from activated carbon,\textsuperscript{14,15} biomaterials,\textsuperscript{16,17} and zeolites.\textsuperscript{18,19–26} Among the many adsorbents, zeolite has good adsorption properties due to its high specific surface area. Therefore, zeolite is considered to be the most common adsorbent.

In addition to the above methods, Saleh\textsuperscript{27} used the synthesis of multiwalled carbon nanotubes and silica nanocomposites (CNT/SiO\(_2\)), and the prepared nanocomposites were evaluated as adsorbents for the removal of Pb(II) from aqueous solutions. The obtained MWCNT/SiO\(_2\) showed a good adsorption performance (~95\%) for silica nanoparticles (~50\%) and carbon nanotubes (~45\%). Basaleh et al.\textsuperscript{22} synthesized polyamide-modified baghouse dust nanocomposites (PMBH) using steel industry waste as the raw material, and the results showed that Pb(II) was adsorbed with a maximum adsorption of 119 mg/g and a maximum removal efficiency of 99.8\%. Jamiu et al.\textsuperscript{23} used N,N-diallyl aspartic acid hydrochloride, 1,1,4,4-tetraallylpiperazine dichloride, and sulfur dioxide as raw materials in the presence of azoisobutyronitrile, and cross-linked poly(amphoteric/anionic) aspartic acid residues were synthesized by the Butler cyclization reaction in the presence of azoisobutyronitrile. EDX analysis confirmed the adsorption of Pb(II) on the polymer. The results indicate that a polymer can be used as an effective adsorbent to adsorb toxic Pb(II) from contaminated aqueous solutions. Hannatu et al.\textsuperscript{24} used ZnO nanoparticles to modify talc to form ZnO—talc nanocomposites (NCs), and the maximum adsorption capacity of this adsorbent for Pb was 48.3 mg/g. Alswat et al.\textsuperscript{25} prepared zeolite—copper oxide (CuO) and zeolite—iron oxide (Fe\(_2\)O\(_3\)) NCs by coprecipitation method, and the solution of zeolite—Fe\(_2\)O\(_3\) NCs prepared with 0.15 g zeolite—Fe\(_2\)O\(_3\) could remove 97.2\% of Pb(II) in 40 min from a 100 mg/L aqueous solution of Pb(II) at a pH between 4 and 6.

In recent years, many researchers have looked to fly ash (FA) solid waste as a raw material to synthesize zeolite adsorbents.\textsuperscript{26} Jessica et al.\textsuperscript{27} produced zeolites by the calcination of FA and a hydrothermal treatment with NaOH. The maximum adsorption capacity for Cu\(^{2+}\) was as high as 142 mg/g, much higher than that of a commercial polymer resin (46.6 mg/g). However, because of the incomplete combustion of coal, FA contains more than 6\% unburned carbon, which cannot be utilized in the synthesis of zeolites.\textsuperscript{28–30} If the FA is activated by the other appropriate methods, excellent zeolite or activated adsorbents can be obtained. Bandura et al.\textsuperscript{31} prepared the zeolite—carbon composites of faujasite and gismondite using high-carbon fly ash (HCFA) as the raw material via a single-step hydrothermal conversion with NaOH, and the results indicated that their adsorption capacity for oil was up to 1.39 g/g. However, this synthesis process produces a large amount of waste alkali that is difficult to recover, which inevitably impedes its wide adoption in sewage treatment. Therefore, the main research goal for converting FA into a sieve adsorbent is to develop a low-cost, efficient, and environmentally friendly adsorbent.

FA is the main solid waste discharged from coal-fired power plants. Its the main components are Si, Al, and other elements. FA is a low-cost, abundant, and easy to obtain precursor of zeolites. The molecular structure of FA is destroyed through chemical reaction under the proper conditions such that the atoms can be rearranged to obtain sieves.\textsuperscript{32,33} If the FA is modified through a chemical reaction under the proper conditions with appropriate methods, it is expected to create excellent adsorbents.

In this paper, FA was mainly composed of Al\(_2\)O\(_3\) (19.8 mass %), SiO\(_2\) (43.4 mass %), and unburned carbon (10.7 mass %) and was used as a raw material to prepare an adsorbent with good performance by a two-step process, which consisted of alkali fusion and a hydrothermal treatment. As a result of the two-step FA hydrothermal treatment, zeolites were obtained on the surface of a honeycomb-structured carbonaceous matrix, which was named a honeycomb-structured activated carbon—zeolite composite (CZC, modified FA). The superficial area of the CZC is six times greater than that of FA, and the adsorption capacity for Pb(II) can reach up to 185.68 mg/g from simulated lead-containing wastewater. In addition, due to the low solubility of inorganic salts in ethanol, the liquid sodium hydroxide waste from the hydrothermal treatment can be recovered by adding ethanol to the mixture. After the distillation of ethanol in sodium hydroxide, the sodium hydroxide can be recycled for alkali fusion, and the ethanol can also be recovered for use. In this scheme, waste alkali is recycled, which not only avoids the secondary pollution of waste alkali to the environment but also reduces the economic cost. This provides a low-cost and effective approach for simultaneously utilizing the C, Si, and Al in FA after the incomplete combustion of coal to prepare a zeolite—activated carbon adsorbent, which has a good adsorption performance for Pb(II) in wastewater. This technology has the advantages of being green and pollution-free in addition to protecting the environment and is expected to realize the high-value utilization of FA.

\section*{Materials and Methods}

\textbf{Preparation of the Composite.} The coal fly ash (CFA) used in this study was obtained from the Midong Electric Power Company Limited (China). A two-step method, alkali fusion and a hydrothermal treatment (i.e., the alkali fusion—trace water method), was used to convert the CFA into a composite of zeolites, while the carbon component of the CFA was carbonized. First, 2.0 g of untreated fly ash was mixed with 4.0 g of NaOH to obtain a homogeneous mixture, then the mixture was heated in a crucible at 750 ℃ (the activation temperature for the general preparation of activated carbon) in a N\(_2\) atmosphere for 1 h. The molten product was then ground and suspended in 24 mL of deionized water, and the mixture was stirred and aged for 2 h at room temperature. Finally, the samples were separated by filtration, washed several times with deionized water (until the pH of the filtrate reached 7), dried at 50 ℃ for 12 h, and used for adsorption experiments.

\textbf{Raw Materials and Reagents.} The lead (Pb) standard solution (1000 μg/mL) was obtained from the National Center for Analysis and Testing of Nonferrous Metals and Electronic Materials. All other reagents were obtained from Tianjin Zhiyuan Chemical Reagent Co.

\textbf{Static Adsorption Experiments of the Composite.} Atomic absorption spectroscopy (AAS) was used for the determination. A 10.0 mL aliquot of the standard solution was
pipetted into a plugged centrifuge tube and adjusted to the appropriate acidity. To the solution was added 0.100 g of the compound, and the plug was closed. The mixture was stirred at a speed of 200 rpm for a certain period of time and then filtered. The filtrate was measured by the FAAS method. The difference subtraction method to calculate the concentrations of Pb²⁺ and CPb in the filtrate Pb²⁺ (qₑ, mg/g). The amount of adsorbed metal ions qₑ (mg/g) was calculated from the following formula:

\[ qₑ = \frac{(Cᵢ - Cₑ)V}{m} \]  

In the formula, \( Cᵢ \) is the initial mass concentration of the metal ion solution (mg/L), \( Cₑ \) is the residual mass concentration of metal ions (mg/L), \( V \) is the volume of the adsorption solution (mL), and \( m \) is the quantity of the adsorbent (g).

The adsorption rate (\( E \)), was calculated as follows:

\[ E = \frac{(C₀ - Cₑ)}{C₀} \times 100\% \]  

where \( C₀ \) is the initial mass concentration of the Pb²⁺ solution (mg/L) and \( Cₑ \) is the equilibrium mass concentration of the Pb²⁺ solution after adsorption (mg/L).

**RESULTS AND DISCUSSION**

**Characterization of Materials.** Figure 1 displays the XRD comparison of FA and CZC. Based on previous work, the XRD pattern of Figure 1a shows that the main phases in FA are silica at 20.8°, 26.7°, 36.5°, 42.4°, 50.1°, 59.9°, and 67.9°, corresponding to the (100), (101), (110), (200), (112), (211), and (212) planes of SiO₂ (JCPDS 01-089-1961), respectively; calcium carbonate at 29.3°, 35.6°, 57.3° and 60.6°, corresponding to the (104), (110), (122), and (214) planes of CaCO₃ (JCPDS 01-072-1937), respectively; and the glass phase of aluminosilicate. It can be seen by comparing Figure 1a and b that the phase of the FA changes obviously after the trace water and alkali dissolution of FA, with the peaks appearing at about 2θ values of 20.9°, 34.5°, 35.6°, 49.4°, and 61.7° corresponding to the (111), (220), (221), (400), and (422) planes of NaAlSiO₄ (JCPDS 01-076-1733), respectively. These were indexed to a zeolite. Combined with the SEM photographs analysis, it was found that there was a large amount of carbon in the CZC; however, there was no obvious peak in the XRD spectrum, indicating that the carbon in CZC was amorphous.

Figure 2 shows the morphological changes (SEM) of FA and the CZC. A comparison between the SEM images of FA and those of the CZC, as seen in Figure 2a and b, shows that the CZC has a larger specific surface area and a distinct pore structure, indicating that the material grew uniformly throughout the preparation process. The pore diameter of its the CZC is about 0.1–0.35 μm, and the zeolites are crystallized and sporadically distributed on the surface of the carbonaceous matrix of the honeycomb structure, indicating that the bulk structure of the raw FA was prepared to form a honeycomb structure.

Figure 3 presents the adsorption and desorption isotherms for FA and the CZC. The adsorption value increased more between the 0.2 and 0.8 relative pressures for CZC that for FA, indicating there are more micropores and mesopores in the CZC. Table 1 shows the porosimetric results for the CZC. An estimated value of 62.5 ± 1.4 m²/g was obtained for the surface area of the CZC, whereas the surface area for FA

![Figure 1. XRD patterns of (a) CZC and (b) FA.](https://example.com/image1)

![Figure 2. SEM photographs of (a and b) CZC and (c and d) FA.](https://example.com/image2)

![Figure 3. N₂ adsorption and desorption BET isotherms for FA and CZC.](https://example.com/image3)
amounted to 11.0 ± 0.8 m²/g. These observations suggest that modifying FA enhanced its isothermal adsorption.

According to the recorded loss on ignition values (TG) and the curves representing the derivatives of the weight loss (DTG), as shown in Figure 4, CZC weight loss occurred according to a three-stage process. In the first stage, weight loss was observed between 50 to 150 °C due to the evaporation of H₂O. The DTA curve can clearly show the change of the water content in the zeolite, and a strong endothermic peak appears near 110 °C, which is the change of bound water in the larger pores of the zeolite. The second stage took place between 150 and 450 °C. The weight loss amounted to ca. 7.8%, and an absorption peak near 420 °C appeared in the DTA curve, which could be intramolecular water removal. The final weight loss was observed between 450 and 700 °C. An exothermic peak appeared at about 600 °C in the DTA curve, corresponding to the crystal transformation of the zeolite with the increase of the temperatures. This was attributed to the dissociation and recombination of silicon and aluminum in Al₂O₃Si₂O₇ to form NaAlSiO₄.

**Langmuir–Freundlich Adsorption Isotherm.** The Langmuir and Freundlich equations are usually used to investigate the adsorption mechanism of adsorbents in aqueous solutions, and the equilibrium isotherm indicates the amount of adsorbate (Pb²⁺) adsorbed by the adsorbent (CZC) in ratio with the equilibrium concentration. In addition, certain phenomena from the adsorbent–adsorbent interaction can be explained by equilibrium curves.

The sorption isotherms curve of Pb²⁺ on CZC (Figure 5) and the Langmuir and Freundlich parameters (Table 2) were identified based on the Langmuir and Freundlich model, and the mathematical model that best represents CZC is the Freundlich model. In this study, the equilibrium isotherms used for CZC were the Langmuir model (R² = 0.957) and the Freundlich model (R² = 0.984), which indicated that the higher the concentration, the greater the adsorption capacity until the available exchangeable sites reach their limit. Therefore, there is a competition for effective exchange sites among the metal ions, and this isotherm shows that the adsorption of Pb²⁺ on CZC is due to relatively weak van der Waals forces.

**Adsorption kinetics.** The static kinetic curves of CZC obtained from the batch experiments (Table 3 and Figure 6) were analyzed using first-order equations and pseudo-second-order equations.

**Table 1. Textural Characteristics of FA and CZC**

| material | S_{BET} (m²/g) | V_p (cm³/g) | A_PD (Å) |
|----------|----------------|-------------|-----------|
| FA       | 11.0 ± 0.8     | 0.03        | 3.4       |
| CZC      | 62.5 ± 1.4     | 0.31        | 12.7      |

*S_{BET} is the specific surface area, V_p is the pore volume, and A_PD is the average pore diameter (calculated via the BET method).
It can be seen in Figure 6 that the adsorption process is divided into two steps. The first step is the rapid adsorption of metal ions within the first 40 min, and the adsorption is a function of the contact time. This is followed by a slower second step in which equilibrium is reached. This behavior, consisting of an initial fast and quantitatively dominant adsorption step followed by a slower and quantitatively insignificant adsorption step, has been widely reported.\(^\text{18,19,25}\)

Combined with the SEM image of the material, the CZC material with a honeycomb-like internal structure has a large number of active sites, which can rapidly adsorb heavy metal ions at the initial stage of adsorption. At a later stage, as more and more active sites are occupied, the adsorption efficiency gradually decreases and, the adsorption slows, and the diffusion from the surface to the internal pores becomes slow and then gradually reaches adsorption equilibrium.

## CONCLUSION

1. This work demonstrated the successful synthesis of a honeycomb-structured zeolite–carbon composite (CZC) by a two-step trace aqueous–alkali fusion and hydrothermal reaction method using FA as the raw material.
2. The sodium hydroxide waste liquid in the hydrothermal treatment can be recovered and recycled by adding ethanol. The product is pollution-free, low cost, environmentally friendly, and simple to operate. The obtained adsorbent CZC proved to be a effective method of removing Pb(II) from aqueous solutions.
3. Compared with FA, the specific surface area of CZC is about six times larger, and the average pore diameter of increased from 3.4 to 12.7 μm.
4. After 40 min of adsorption at pH 7, the CZCs showed an uptake of 185.68 mg/g Pb(II). The kinetics studies showed that pseudo-second-order kinetics is more consistent with the adsorption of Pb(II) ions onto the CZC surface.
5. Compared to other FA-derived adsorbents, the CZC showed a higher adsorption capacity. Therefore, it has good potential to be applied as an adsorbent for mitigating wastewater pollutants.

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### Author Contributions

M.Z. was responsible for investigation, experimental design, data analysis, and writing part of the original draft. D.C. was responsible for instrumentation operation. Y.L. was responsible for supervision and editing.

### Notes

The authors declare no competing financial interest. Some or all data used during the study are available in a repository or online in accordance with data retention policies.

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