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Original Paper

Phosphorus Tailings Modified Biochar for the Removal of Phosphorus from Water

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Abstract

Dehydrated sludge and phosphate tailings are used as raw materials, mixed in a certain proportion, and pyrolyzed in a tube furnace at a temperature of 800 ℃ to form sludge composite phosphate tailings biochar, which is then used for the treatment of phosphate containing wastewater. We investigated the adsorption efficiency and influencing factors (initial phosphorus concentration, pH value, and coexisting ions) of biochar for phosphorus in both single and mixed systems (coexistence of phosphorus, ammonia nitrogen, and paracetamol). The results showed that in a single system, when the phosphorus concentration was 50mg/L, the pH value was 7, and the concentration of coexisting ion SO_4^{2-} was 0.1mg/L, the removal rates were 99.88%, 80.96%, and 86.04%, respectively. Biochar had the best adsorption effect on phosphorus. In the mixed system, the removal rate was 99.86% when the phosphorus concentration was 50mg/L, and the removal rates were 99.86%, 54.98%, and 79.10% when the coexisting ion SO_4^{2-} concentration was 0.1mg/L at pH 8. The adsorption effect on phosphorus was the best. Phosphorus is one of the main elements causing eutrophication of water bodies, which can lead to the proliferation of algae in the water. So the removal of phosphorus from water is very important.

Keywords

Biochar, phosphorus tailings, pH, phosphate, co-existing ions

1. Introduction

Due to the increase in domestic sewage volume, a large amount of organic and inorganic phosphorus enters the water body, causing a significant proliferation of algae in the water, and ultimately leading to eutrophication of the water body.Therefore, removing phosphorus from wastewater becomes an essential step in wastewater treatment. Among the many methods for treating phosphorus in wastewater, the adsorption method has received widespread attention due to its good treatment effect, low cost, and low secondary pollution (Huang, Li, Hou et al., 2024). Natural minerals, biomass materials, metal oxides, and several emerging types of adsorbents (Liu, Wang, Chen et al., 2025). Biochar has attracted much attention due to its wide raw material sources, well-developed pores, abundant surface functional groups, as well as its simple preparation process and low cost (Mi, Liu, Huang et al., 2025). These characteristics make biochar an ideal material for water pollution control. Since sludge is mainly composed of water, organic matter, microorganisms, inorganic particles and heavy metals, the thermal decomposition of organic matter can produce biochar, achieving the resource utilization of solid waste. Phosphorus tailings are the solid waste remaining after the beneficiation process of phosphorite. Their main components include unused phosphorite minerals, quartz, clay minerals, etc. The main components of them are $CaCO_3$ and $CaMg(CO_3)_2$. If they are stored in the environment for a long time, they may cause environmental problems such as water pollution and soil degradation (Ma, Li, Yue et al., 2025). The pore structure and surface functional groups of unmodified sludge biochar have certain limitations. By modifying the biochar, efficient adsorption of pollutants can be achieved (Liu, Chen, Li et al., 2025). Studies have shown that the appropriate addition of phosphorus tailings can increase the organic carbon content of sludge biochar, while also enhancing its surface activity and adsorption properties (Yang, Lv, Zhou et al., 2022). The introduction of phosphorus tailings can not only increase the functional group content in biochar, but also introduce metal elements such as Ca and Mg. Through precipitation, pollutants can be removed. The main content of this experiment is to study the removal effect of sludge-combined phosphorus tailings biochar on phosphorus in water. In the dehydrated sludge, a certain proportion of phosphorus tailings was added, and the sludge composite phosphorus tailings biochar was prepared through pyrolysis at a temperature of 800°C. By comparing the adsorption effects of phosphate on different concentrations of phosphorus-containing solutions, different pH values, and different concentrations of coexisting ions, the optimal adsorption environment was identified. This provides a scientific basis for the removal of phosphorus from wastewater and the disposal of sludge and phosphorus tailings.

2. Materials and Methods

2.1 Experimental Materials

The sludge and phosphorus tailings used in this experiment were respectively sourced from the dewatering sludge of a wastewater treatment plant in Guiyang City and a phosphorus mine in Guizhou Province. After collecting the sludge, it is mixed with the phosphorus tailings and then undergoes natural air-drying, grinding and pulverizing processes. The resulting powder is dried at 105°C until its mass reaches a constant state. It is then sieved through a 100-mesh screen and stored for future use.

2.2 Experimental Reagents and Instruments

The main reagents used in the experiment are listed in Table 1. Potassium dihydrogen phosphate and ammonium chloride are of analytical purity of grade A, while other reagents are of analytical purity greater than 98%. The experimental water is deionized water. The main instruments used in this experiment are shown in Table 2.

Table 1. Reagents Used in the Experiment

Reagent Name	Chemical formula	Specification	Manufacturer
Potassium dihydrogen phosphate	KH_2PO_4	500g	Tianjin Youping Chemical Reagent Co., Ltd.
Ascorbic acid	$C_6H_8O_6$	25g	Tianjin Kemioco Chemical Reagent Co., Ltd.
Ammonium molybdate	$(NH_4)_2MoO_4$	500g	Chongqing Beibei Chemical Reagent Co., Ltd.
Potassium stibate tartrate	$C_8H_4K_2O_{12}Sb_2$	500g	Tianjin Kemioco Chemical Reagent Co., Ltd.
Sulfuric acid	H_2SO_4	500mL	Chuan Dong Chemical Industry
Ammonium chloride	NH_4Cl	500g	Tianjin Kemioco Chemical Reagent Co., Ltd.
Acetaminophen	$C_8H_9NO_2$	25g	Shanghai Aladdin Biochemical Technology Co., Ltd.
Potassium sulfate	K_2SO_4	500g	Chengdu Jinshan Chemical Reagent Co., Ltd.

Table 2. Main Instruments and Equipment

Equipment Name	Model number	Manufacturer
Ultraviolet-visible spectrophotometer	UV-5500	Shanghai Yuanxie Instrument Co., Ltd.
Constant temperature water bath oscillator	THZ-82	Jinan Uike Experimental Equipment Co., Ltd.
pH meter	P301	Shanghai Youke Instrument and Meter Co., Ltd.
Tube-type electric furnace	SK-G06123K	Tianjin Zhonghuan Electric Furnace Co., Ltd.

Electronic analytical balance	JF2104	Yuyao Jinnuo Tianping Instrument Co., Ltd.
Electrically heated constant-temperature drying oven	202-2AB	Tianjin Taistel Instrument Co., Ltd.
Ultra-pure water machine	WP-VP-IV-30	Sichuan Wouter Water Treatment Equipment Company

2.3 Preparation of Sludge-combined Phosphorus Tailings Biochar

Using the remaining sludge after sieving and the phosphorus tailings as raw materials, mix the phosphorus tailings with the remaining sludge at a mass ratio of 1% evenly, and then put them into a tubular furnace. Before thermal decomposition, it is necessary to first introduce 30 minutes of 1L/min nitrogen gas, and then increase the temperature at a rate of 5℃/min to 800℃ for thermal decomposition for 2 hours. After cooling to room temperature, take it out and store it in a drying container.

2.4 Adsorption Experiment of Phosphorus by Sludge Composite Phosphate Tailings Biochar

0.1g of the sludge composite phosphorus tailings biochar was weighed and added to 50 mL of a single or mixed solution (with an ammonia concentration of 25 mg/L and a paracetamol concentration of 20 mg/L) containing 100 mg/L of phosphorus. The solution was placed in a constant temperature water bath shaker and shaken for 24 hours. Then it was taken out and filtered through non-phosphorus filter paper. The absorbance of the solution was measured using a UV spectrophotometer, and the concentration of the remaining phosphorus solution in the solution was calculated. This study conducted three parallel experiments, and the results were averaged.

2.5 Factors Affecting Adsorption

Initial Phosphorus Concentration

An adsorption experiment was conducted using biochar from sludge combined with phosphorus tailings at a temperature of 800℃. The influence of single phosphorus solutions and mixed solutions with different initial phosphorus concentrations (50mg/L, 75mg/L, 100mg/L, 125mg/L, 150mg/L) on the adsorption of the biochar was screened through this experiment. Other experimental conditions were the same as those of the adsorption experiment.

pH

The pH values of the single phosphorus solution and the mixed solution were adjusted to 3, 4, 5, 6, 7, 8, 9, 10, and 11 respectively. The influence of pH on adsorption was studied through experiments, with other experimental conditions remaining the same as those of the adsorption experiment.

Coexisting ions (SO_4^{2-})

By preparing single phosphorus solutions and mixed solutions with different concentrations of coexisting ions (0.001mg/L, 0.01mg/L, 0.1mg/L), the influence of coexisting ions on the adsorption experiment was studied. Other experimental conditions were the same as those of the adsorption experiment.

2.6 Result Calculation

The calculation methods for the equilibrium adsorption capacity (qe) and removal rate (E) of phosphorus by biochar are as follows:

$$Q = \frac{(C_0 - C)V}{M} \tag{1}$$

$$E = \frac{C_0 - C}{C} \times 100\% \tag{2}$$

In the formula:

qe - equilibrium adsorption amount, mg/g;

C0 - initial phosphorus mass concentration, mg/L;

C - phosphorus mass concentration at adsorption equilibrium, mg/L;

V - volume of the solution, L;

m - mass of the added biochar, g;

E - removal efficiency, %.

3. Results and Discussion

3.1 The Influence of Initial Phosphorus Concentration on Adsorption

Solution of a single pollutant

Figure 1 shows the adsorption of phosphorus by the sludge composite phosphorus tailings biochar under different initial phosphorus concentrations. As can be seen from the figure, as the phosphorus concentration increases, the removal rate of phosphorus by the sludge biochar shows a downward trend. When the initial phosphorus concentration was 50mg/L, the removal rate of phosphorus by the sludge biochar reached its maximum of 99.88%. When the initial phosphorus concentration increased to 150mg/L, the removal rate of phosphorus by the sludge biochar was 49.23%.

The higher the initial phosphorus concentration, the more conducive it is to adsorption. When the phosphorus concentration is 50mg/L, the adsorption amount is 24.95mg/g, and when it is 150mg/L, the adsorption amount is 37.03mg/g. From this, it can be seen that the higher the initial concentration of phosphorus, the more phosphorus is adsorbed. This is because at low concentrations, the adsorption sites of biochar are not fully occupied, so the removal rate is high and the adsorption amount is low. When the concentration is high, the adsorption sites will be completely occupied, and there are still some phosphorus molecules in the solution that have not been fully adsorbed. Therefore, the removal rate is low while the adsorption capacity is high. In the experiment of phosphate adsorption on modified rice straw biochar prepared by co pyrolysis of magnesium oxide, the adsorption amount showed an upward trend with the increase of phosphorus concentration, which is similar to the results of this study[7].

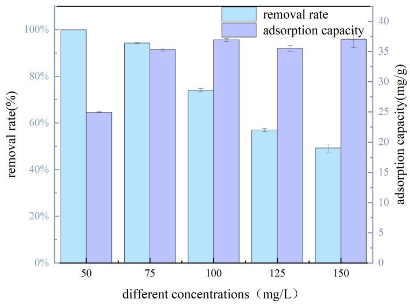

Figure 1. Initial Phosphorus Concentration in the Solution of a Single Pollutant

Solution of mixed pollutants

In the solution containing mixed pollutants, the influence of different initial phosphorus concentrations on the adsorption effect of sludge composite phosphorus tailings biochar is shown in Figure 2. As shown in the figure, in the solution containing mixed pollutants, the adsorption of phosphorus on the biochar is similar to that in the solution with a single pollutant. The removal rate decreases as the concentration of phosphorus in the solution increases. Among them, in the mixed solution with a phosphorus concentration of 50mg/L, the removal rate reaches the highest at 99.87%, which is very close to the removal rate in the solution with a single pollutant. When the initial phosphorus concentration is low, the ammonia nitrogen and paracetamol in the mixed solution have little effect on the phosphorus adsorption by the modified biochar. This might be because at low concentrations, almost all the phosphorus in the solution is adsorbed, but the sites of the biochar have not been fully occupied.At a concentration of 150mg/L, the removal rate of phosphorus by the sludge biochar was 42.06%. When the phosphorus concentration was high, the adsorption of ammonia nitrogen and paracetamol in the mixed solution had a certain impact on phosphorus. Because the sludge biochar not only adsorbed phosphorus but also ammonia nitrogen and paracetamol, some of the adsorption sites were occupied by ammonia nitrogen and paracetamol, resulting in a decrease in the phosphorus adsorption rate.

When the initial phosphorus concentration was 50 mg/L, the adsorption amount was 24.95 mg/g. The adsorption amount showed an upward trend as the concentration increased, which was similar to the situation of the adsorption of a single pollutant. At low concentrations, the adsorption sites of biochar were not fully occupied by phosphorus, ammonia nitrogen and paracetamol. Therefore, almost all the phosphorus in the solution was absorbed, resulting in a high removal rate and a low adsorption capacity. At high concentrations, all the adsorption sites in the biochar are occupied, and there is still some unadsorbed phosphorus remaining in the solution. Therefore, the removal rate is low and the adsorption capacity is high. Moreover, ammonia nitrogen and paracetamol will occupy some of the sites in the biochar, resulting in a lower adsorption capacity compared to a single solution.

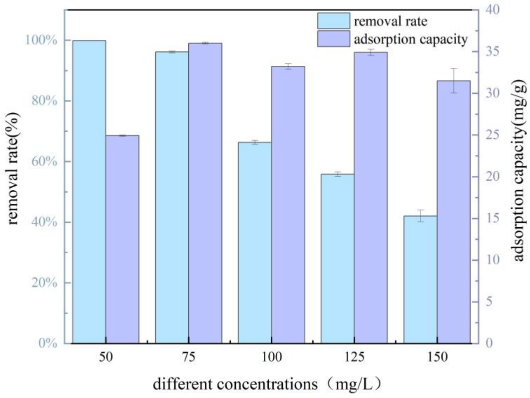

Figure 2. Solutions of Mixed Pollutants with Different Initial Phosphorus Concentrations

The influence of pH value on adsorption

Solution of a single pollutant

The pH value of the solution is one of the key factors influencing the adsorption process. Different pH ranges will lead to significant differences in the forms of phosphate, thereby affecting the adsorption effect of biochar. The existence form of phosphate varies significantly with the change of solution pH value, showing distinct interval characteristics. Within the pH range of 2 - 7, phosphate mainly exists in the form of H_2PO^{4-}. When the pH value rises to 7 - 12, it mainly transforms into the form of HPO_4^{2-}. Due to the higher adsorption energy of HPO_4^{2-} compared to H_2PO^{4-}, its adsorption difficulty significantly increases. This results in a downward trend of adsorption capacity as the solution pH value rises[8].

As shown in Figure 3, in solutions with pH ranging from 3 to 11, the removal rate of phosphorus by sludge biochar was within the range of 71.04% to 80.96%.When the pH value was 7, the removal rate of phosphorus by the sludge biochar was the highest, reaching 80.96%. Under neutral conditions, the removal rate of phosphorus was the highest. In acidic conditions, the removal rate increased with the increase of pH, while in alkaline conditions, the opposite was true.

The adsorption amount of phosphorus shows an upward trend within the pH range of 3 to 7, and a downward trend within the range of 7 to 11. The pH value of the solution has a significant impact on the surface properties of biochar and the form of phosphorus present. Under acidic conditions, as the pH value of the solution increases, the existence form of phosphorus gradually changes from $H_2PO_4^-$ and HPO_4^{2-} to HPO_4^{2-} and PO_4^{3-}. This results in an increase in the negative charge of phosphorus, thereby enhancing its electrostatic attraction to the positively charged surface of the activated sludge biochar, ultimately increasing the adsorption capacity of phosphorus. Under alkaline conditions, the OH- ions in the solution will compete with PO_4^{3-} for adsorption sites, resulting in a decrease in the amount of phosphorus adsorbed (Tu, Zhang, Cen et al., 2023). This trend of change is the same as that of the modified corn husk biochar's adsorption of phosphorus under different pH values. In all cases, the removal rate of phosphorus from water is the highest under neutral conditions, which is similar to the results of this study. When the concentration is high, the adsorption sites will be completely occupied, and there are still some phosphorus molecules in the solution that have not been fully adsorbed. Therefore, the removal rate is low while the adsorption capacity is high (Liu & Yue, 2024).

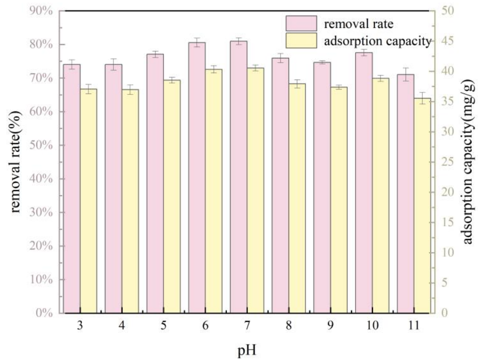

Figure 3. Solutions of a Single Pollutant at Different pH Values

Solution of mixed pollutants

In the mixed pollutant solution with a phosphorus concentration of 100 mg/L, the influence of pH changes on the adsorption of phosphorus by the sludge composite phosphorus tailings biochar is shown in Figure 4. From the trend of the removal rate in the figure, it can be seen that pH has a significant impact on the adsorption of phosphorus by the biochar. The removal rate is higher in a neutral environment, and the adsorption capacity of phosphorus is also the highest. At a pH of 8, the removal rate was the highest, reaching 54.98%.Under acidic conditions, when multiple pollutants coexist, the removal rate of phosphorus decreases significantly. This is because when the pH value is low, the H^+ ions in the solution will compete with ammonia nitrogen and paracetamol (Wu, Quan, Chen et al., 2024). This resulted in the fact that the H^+ ions that should have been adsorbed in the solution were not adsorbed. The negative charge of phosphorus in this solution was less than that in the single solution, so the adsorption amount decreased significantly compared to the single solution.

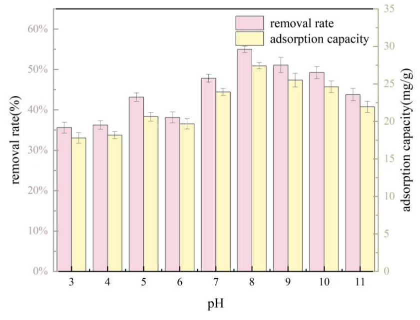

Figure 4. Solutions of Mixed Pollutants at different pH values

The influence of coexisting ions on adsorption

The factors that can affect the adsorption efficiency of biochar are complex and diverse, including pyrolysis conditions, physical and chemical properties, pH value of the solution, coexisting ions, and characteristics of the pollutants (Wu, Quan, Chen et al., 2024). The coexisting anions in the actual wastewater will compete with phosphate for adsorption sites, thereby reducing the phosphorus removal efficiency of the biochar (Wu, Quan, Chen et al., 2024). Since SO_4^{2-} is a common anion in natural water bodies and both PO_4^{3-} and SO_4^{2-} can interact with the surface active sites of biochar in water and compete for the adsorption sites, this is the case. Therefore, in this study, SO_4^{2-} was taken as the example. The removal rate of phosphorus by the sludge composite phosphorus tailings biochar was investigated under the presence of different concentrations of coexisting ions.

Solution of a single pollutant

As shown in Figure 5, in the solution with a phosphorus concentration of 100 mg/L, when the concentration of SO_4^{2-} was 0.001 mg/L, 0.01 mg/L, and 0.1 mg/L, the removal rate of phosphorus by the sludge biochar was 84.34%, 82.59%, and 86.04% respectively, showing a trend of first decreasing and then increasing. At a concentration of 0.1mg/L, the removal rate is the highest. This might be because when SO42- combines with phosphate, the coagulation reaction becomes more intense (Ma, Zhao, Pan et al., 2025). The removal rate of phosphorus in the blank control was 82.24%. After the addition of coexisting ions, the removal rates increased by 2.10%, 0.35%, and 3.80% respectively, which promoted the adsorption of sludge biochar. However, the degree of promotion was very small, indicating that the adsorption competition of phosphate and SO_4^{2-} on the surface of biochar was relatively weak. The conclusion of the experiment on the adsorption of phosphates by dehydrated sludge and dolomite-modified biochar regarding the influence of coexisting ions is the same. The influence of SO42- on the adsorption effect is not significant (Li, Li, Huang et al., 2019).

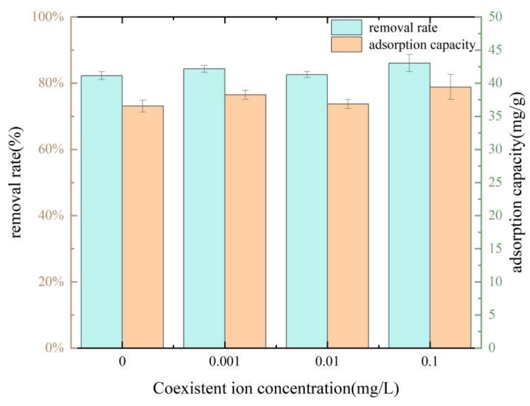

Figure 5. Concentrations of Different Coexisting Ions (SO_4^{2-}) in the Solution of a Single Pollutant

Solution of mixed pollutants

Figure 6 shows the influence of the presence of different concentrations of coexisting ions in a mixed pollutant solution with a phosphorus concentration of 100 mg/L on the adsorption effect of sludge biochar. As can be seen from the figure, the trend of the removal rate is the same as that in the single pollutant solution, both showing a trend of first decreasing and then increasing. However, the removal rate is relatively low, being 56.25%, 42.44%, and 79.10% respectively. This indicates that the ammonia nitrogen, paracetamol and SO_4^{2-} in the mixed solution compete for adsorption sites, resulting in a decrease in removal rate.

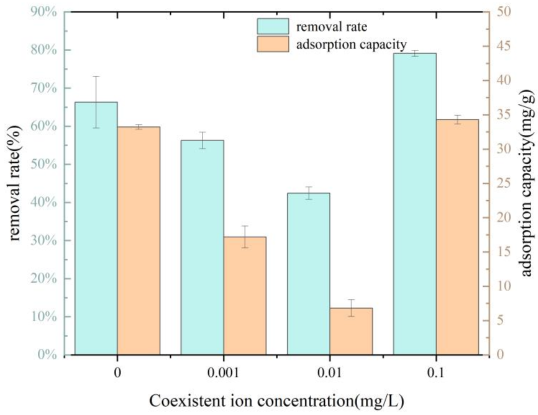

Figure 6. Different Concentrations of Coexisting Ions (SO_4^{2-}) in a Solution of Mixed Pollutants

4. Conclusion

(1) Different initial phosphorus concentrations have certain effects on the sludge composite phosphorus tailings biochar. The removal rate decreases as the initial phosphorus concentration increases. When the initial phosphorus concentration was 100mg/L, the ammonia nitrogen and paracetamol in the solution had a significant impact on the removal rate of phosphate, reducing it by 7.7%. At other initial concentrations, the ammonia nitrogen and paracetamol in the solution had a relatively minor impact on the removal rate of phosphate.

(2) In solutions with the same pH, the sludge composite phosphorus tailings biochar also has an impact on the removal of phosphate. The results show that in both single-pollutant solutions and mixed-pollutant solutions, a neutral environment is conducive to the removal of phosphate. In the environment with ammonia nitrogen and paracetamol, the removal rate of biochar will be significantly reduced.

(3) With the presence of the coexisting ion SO_4^{2-}, the removal rate of biochar in the single pollutant solution shows a certain increase, but the overall impact is relatively small. In the mixed pollutant solution, the influence of coexisting ions is also the same. However, due to the presence of other pollutants, the amount of phosphate adsorbed by the sludge biochar is relatively small.

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