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STUDY ON THE ADSORPTION PERFORMANCE AND MECHANISM OF PB (II) IN WATER BY BIOCHAR PREPARED FROM COCONUT SHELL

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ABSTRACT

The direct discharge of lead containing wastewater into the environment without treatment poses a serious threat to the ecosystem and human health. Therefore, efficient, environmentally friendly, and low-carbon treatment of lead-containing wastewater is one of the critical issues that need to be addressed urgently. In this study, three types of biochar (designated as B450, B550, and B650) were independently prepared from agricultural and forestry waste coconut shells, and the effects of factors such as adsorption time, initial solution concentration, solution pH, and biochar dosage on the adsorption of Pb(II) in water by biochar were investigated through static adsorption experiments. The experiments showed that at a temperature of 30 °C, an initial solution concentration of 100 mg/L, a dosage of 8 g/L, a pH of 5, and an adsorption time of 3 h, biochar B650 exhibited the best adsorption effect on Pb(II) in water. The isothermal adsorption curves of the three types of biochar on Pb(II) in water could be well fitted with both Freundlich and Langmuir equations, and B650 showed the strongest adsorption capacity for Pb(II) in water.

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INTRODUCTION

Lead (Pb), one of the "five toxic" heavy metals that pollute the ecological environment (Becker, 2022), has a strong ability to migrate and transform. It enters the human environment through a variety of activities, such as mining, metal smelting, automobile exhaust, paint coatings, use of lead-containing pharmaceuticals and chemicals, and production of lead-acid batteries, posing a risk to human health (Siyuan, 2022). Lead and its related compounds can enter the human body through the respiratory tract, digestive system, and skin absorption processes, causing damage to physiological processes and multiple organs. Lead poisoning can cause cirrhosis of the liver, necrosis of the liver, decreased kidney function, and impaired fertility. In addition, lead can pass through the placenta to the fetus, causing fetal malformations or even miscarriages (Xu, 2008). Current treatment methods for lead-containing wastewater include adsorption, membrane separation, ion exchange, and biological treatment, each with its own advantages and limitations in terms of operating costs, treatment costs, and efficiency (Xiutao, 2021). Biochar is a class of insoluble, carbon-rich solid materials produced by the pyrolysis of biomass materials under high temperature and limited oxygen conditions. It has numerous advantages, such as being porous, having a high specific surface area, and being environmentally friendly. In addition, the raw materials for biochar production are widely available, and its adsorption capacity for some heavy metals even exceeds that of activated carbon, which has attracted the interest of many researchers in the field of heavy metal pollution remediation. The adsorption performance of biochar is influenced by its structure. Studies have shown that there is an inverse relationship between biochar particle size and specific surface area; the smaller the particle size, the larger the specific surface area and the more significant the adsorption effect (Hao, 2023). The physicochemical properties of biochar are affected by pyrolysis temperature. In general, the yield decreases with increasing pyrolysis temperature, while other properties such as ash content, specific surface area, and porosity increase with increasing temperature (Dingmei, 2012 and Xinyu, 2021). As one of the typical regional economic crops in tropical areas, coconut shells, which are the main by-product of the coconut food processing industry, are considered a promising agricultural and forestry waste biomass resource with great potential for development due to their low cost and abundant availability. However, there are relatively few reports on the preparation of biochar from coconut shell waste and its application in heavy metal adsorption (Yingchao, 2015; Yingshuian, 2012 and Nan, 2017). Therefore, this study aims to independently prepare biochar from widely available coconut shells in Hainan under different temperature conditions and investigate its adsorption effect on Pb(II) in water to explore the optimal adsorption conditions and mechanisms. This will provide a reliable theoretical basis and scientific foundation for the future treatment of lead-containing wastewater.

MATERIALS AND METHODS

Preparation of Coconut Shell Biochar: Local green coconut shells from Hainan were used in this experiment. The shells were cut into squares of approximately 1 cm side length, placed in a 100 mL crucible, wrapped in aluminum foil, and heated to produce biochar. The biochar was prepared using a limited oxygen programmed heating carbonization method: first, the temperature was raised to 330 °C within 30 minutes and held constant for 60 minutes; then, the temperature was raised to the target temperature (450 °C, 550 °C, 650 °C) at a rate of 10 °C/min and held constant for another 150 minutes; finally, the biochar was allowed to cool naturally to room temperature. The biochar was then removed, ground with a mortar and pestle, sieved through a standard 100-mesh sieve, mixed evenly, and stored in clean, dry, wide-mouth bottles for later use, labeled B450, B550, and B650.

Instruments: ICP Inductively Coupled Plasma Emission Spectrometer (ICAP6300); Muffle Furnace (JHZ-10-12); Programmable Temperature Controller (AI-708P); Electronic Precision Balance (GH-200); Diaphragm Vacuum Pump (GM-1.0A); Constant Temperature Incubator Shaker.

EXPERIMENTAL METHODS

Effect of Different Initial Pb(II) Concentrations on Adsorption Performance: 0.2 g of coconut shell biochar prepared at three different temperatures were weighed and added to 25 mL of Pb(II) solution with concentrations of 10, 20, 40, 60, 80, 100, 120, and 140 mg/L, respectively. The pH was adjusted to 2.0±0.1 and the samples were placed in a constant temperature shaker set at 30 °C and 300 rpm for 3 h. The samples were then filtered through a 0.45 μm membrane filter, and the residual concentration of Pb(II) in the filtrate was measured to investigate the effect of different initial Pb(II) concentrations on the removal efficiency.

The adsorption capacity q was used to evaluate the adsorption performance of biochar for Pb(II) in solution, as shown in equation (1):

$$q = \frac{(C_0 - C_t)V}{M} \qquad \text{......................(1)}$$

Where q is the adsorption capacity of biochar for Pb(II) at time t (mg/g); C_0 is the initial concentration of Pb(II) in the solution (mg/L); C_t is the concentration of Pb(II) in the solution at time t (mg/L); V is the volume of Pb(II) solution added (mL); M is the mass of biochar added (g).

The adsorption isotherms of Pb(II) on coconut shell biochar prepared at three different temperatures at 30 °C were fitted using the Freundlich model (equation (2)) and the Langmuir model (equation (3)).

$$\lg q_e = \lg K_F + \frac{1}{n}\lg C_e \qquad \text{........................(2)}$$

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{K_L q_m C_m} \qquad \text{..........................(3)}$$

Where q_e is the equilibrium adsorption capacity (mg/g); C_e is the equilibrium adsorption concentration of Pb(II)(mg/L); K_F and n are temperature-dependent adsorption characteristic constants, where K_F is in units of ($mg^{1-n} \cdot L^n/g$); q_m is the maximum adsorption capacity (mg/g); K_L is the Langmuir adsorption coefficient (L/mg).

Biochar Dosing Experiment: 0.1, 0.2, 0.3, 0.4, and 0.5 g of coconut shell biochar prepared at three different temperatures were weighed and added to 25 mL of 100 mg/L Pb(II) solution. The pH was adjusted to 2.0±0.1 and the samples were placed in a constant temperature shaker set at 30 °C and 300 rpm for 3 h. The samples were then filtered through a 0.45 μm membrane filter, and the residual concentration of Pb(II) in the filtrate was measured to investigate the effect of different biochar dosages on the removal efficiency of Pb(II).

Effect of Different pH Values on Adsorption Performance: 0.2 g of coconut shell biochar prepared at three different temperatures was weighed and added to 25 mL of 100 mg/L Pb(II) solution. The pH was adjusted to 1, 2, 3, 4, 5, and 6, respectively, and the samples were placed in a constant temperature shaker set at 30 °C and 300 rpm for 3 h. The samples were then filtered through a 0.45 μm membrane filter, and the residual concentration of Pb(II) in the filtrate was measured to investigate the effect of different pH values on the removal efficiency of Pb(II).

Effect of Adsorption Time on Adsorption Performance: 0.2 g of coconut shell biochar prepared at three different temperatures was weighed and added to 25 mL of 100 mg/L Pb(II) solution. The pH was adjusted to 2.0±0.1 and the samples were placed in a constant temperature shaker set at 30 °C and 300 rpm for 3 h. The samples were then filtered through a 0.45 μm membrane filter, and the residual concentration of Pb(II) in the filtrate was measured to investigate the effect of different adsorption times on the removal efficiency of Pb(II).

Statistical Analysis of Data: All experiments were performed in triplicate, and data were obtained by averaging the results of the three experiments. Experimental data were processed using Origin 9.0 and Excel 2020 for statistical analysis.

RESULTS AND DISCUSSION

Influence of different initial Pb(II) concentrations on the adsorption of Pb(II) in water by biochar prepared at different temperatures and their isothermal adsorption characteristics: The influence of different initial Pb(II) concentrations on the adsorption capacity of the three biochars is shown in Figure 1. It can be seen that the adsorption capacities of B450, B550 and B650 for Pb(II) in water increase with the increase of the initial solution concentration and eventually tend to equilibrate. When the amount of coconut shell biochar added is constant, the number of adsorption sites provided is also constant. At lower Pb(II) concentrations, most of the Pb(II) can interact with the adsorption sites on the surface of the adsorbent by attracting and binding to each other. When the initial concentration of Pb(II) is low, the adsorption capacity of biochar increases with the increase of the initial concentration of Pb(II). When the concentration of Pb(II) is high, the adsorption sites of the adsorbent are occupied and the adsorption has reached saturation.

There are not enough adsorption sites to adsorb Pb(II) in the solution, so even if the solution concentration is increased, the adsorption capacity will not increase and may even decrease slightly. Among them, under the condition of an initial concentration of 100 mg/L of Pb(II) solution, the maximum adsorption capacities of B450, B550, and B650 for Pb(II) in water are 6.400 mg/g, 6.538 mg/g, and 6.988 mg/g, respectively. Considering all factors, when the initial solution concentration is 100 mg/L, B650 has the best adsorption effect on Pb(II) with other conditions being constant. At 30 °C, the results of fitting the adsorption isotherms of Pb(II) with Freundlich and Langmuir models for the coconut shell biochar prepared at three different temperatures are shown in Figures 2a and 2b, respectively. According to the above equations (2) and (3), the fitting parameters for the isothermal adsorption of Pb(II) by the three biochars were obtained, and the results are shown in Table 1.

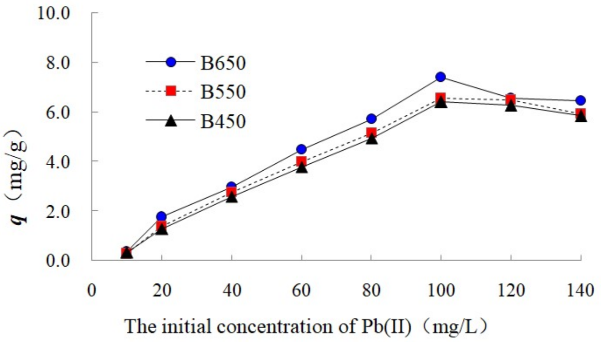

Figure 1. Effect of different initial concentrations on the adsorption of Pb(II)

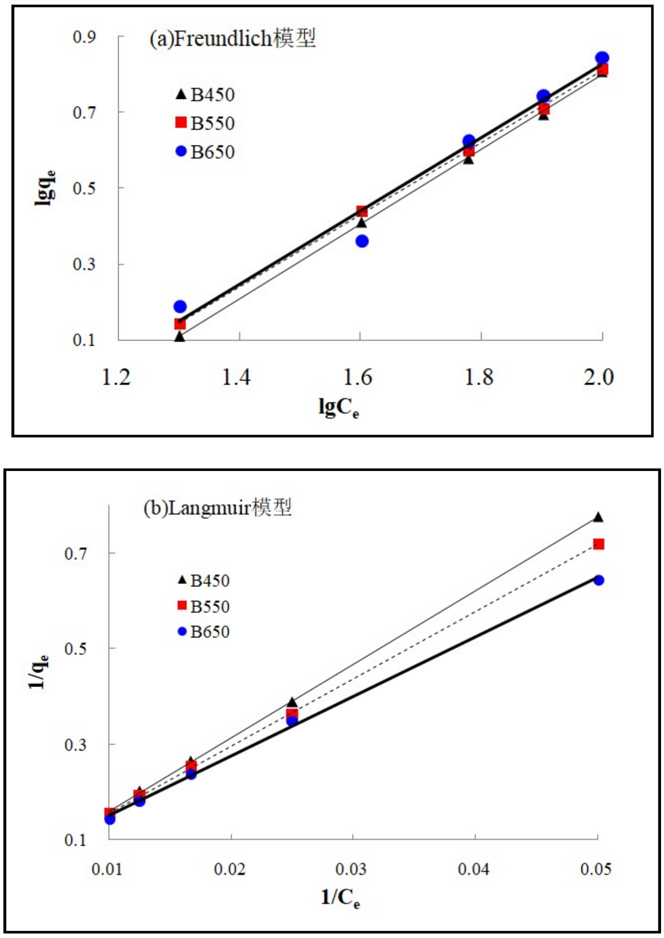

Figure 2. Isothermal adsorption curves of Pb(II) in water by three types of biochar

Table 1. Isotherm adsorption curve fitting parameters

Biochar	Freundlich equation			Langmuir equation		
	$\lg K_F$	$1/n$	r^2	K_L(L/mg)	Q_m(mg /g)	r^2
B450	1.717	0.985	0.9990	0.002	36.630	0.9986
B550	1.094	0.925	0.9990	0.001	67.568	0.9998
B650	1.107	0.966	0.9710	0.004	169.492	0.9998

According to the calculated fitting correlation coefficients (r^2) of the isothermal adsorption equations, both Freundlich and Langmuir equations have good fitting effects on the data of Pb(II) adsorption by the three biochars. The r^2 values are 0.9990, 0.9990, 0.9710, and 0.9986, 0.9998, 0.9998, respectively. Therefore, the adsorption behavior of the three biochars toward Pb(II) is suitable for fitting with both Freundlich and Langmuir equations. From the fitting results of the Freundlich equation, it can be seen that Pb(II) in water can be well adsorbed by all three biochars. According to the fitting of the Langmuir equation, the maximum adsorption capacities of B650, B550, and B450 for Pb(II) are 169.492 mg/g, 67.568 mg/g, and 36.630 mg/g, respectively. Therefore, B650 shows a strong adsorption capacity for Pb(II). At the same time, as shown in Table 1, the adsorption strength (1/n) of the three biochars for Pb(II) is between 0.925 and 0.985. According to the relationship between the value of 1/n and the shape of the adsorption isotherm, the isothermal adsorption strength 1/n of the three biochars for Pb(II) is less than 1, which all belong to the Langmuir adsorption isotherm. This indicates that the three biochars have a strong affinity for Pb(II) at the beginning of isothermal adsorption.

Effect of different biochar dosing on the adsorption of Pb(II) in water: The effect of different addition amounts of biochar on the adsorption capacity is shown in Figure 3. The adsorption capacity of the three biochars for Pb(II) in water first increases and then decreases with the increase in addition amount. In general, the adsorption capacity of biochar will increase as the amount added increases. However, more is not always better; the optimal adsorption effect occurs when the adsorption reaches equilibrium. If the amount of biochar added is too high, the migration of Pb(II) to the biochar will be hindered, resulting in a decrease in the adsorption capacity. When the addition amount is 0.2 g (8 g/L), the maximum adsorption capacities of B450, B550, and B650 are 5.688 mg/g, 5.838 mg/g, and 5.888 mg/g, respectively. Considering all factors, under other constant conditions, the biochar under B650 has the best adsorption effect on Pb(II) in water when the addition amount is 0.2 g.

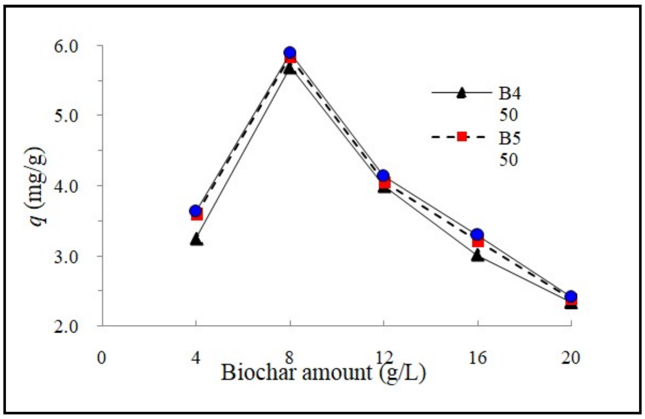

Figure 3. Effect of biochar dosing on the adsorption of Pb(II)

Effect of different pH values on the adsorption of Pb(II): The effect of different pH values on the adsorption of Pb(II) by biochar is shown in Figure 4. The adsorption capacity of the three biochars for Pb(II) in water first increases and then decreases with increasing solution pH. At very low pH values, the biochar surface is dominated by cations, which repel heavy metal ions and result in weaker adsorption. Increasing the acidity of the solution causes an increase in the number of anions on the outer surface of the biochar, thereby increasing the adsorption of metal ions. Both acidic and alkaline conditions are unfavorable for adsorption of heavy metal ions by biochar [41]. When pH = 5, the adsorption capacities of the three biochars reach their maximum values, which are 12.425 mg/g, 12.488 mg/g and 12.513 mg/g, respectively. Considering all factors, when other conditions are constant and pH is 5, the maximum adsorption capacity of biochar under B650 is 12.513 mg/g. Under these conditions, biochar has the best adsorption effect on Pb(II) in water.

Effect of adsorption time on the adsorption of Pb(II) in water by biochar: As shown in Figure 5, the adsorption capacity of the three biochars for Pb(II) in water first increases with time, then remains almost constant, and finally reaches adsorption equilibrium. When the adsorption time is before 3 h, the adsorption capacity gradually increases with time. After 3 h, the increase in adsorption capacity becomes insignificant, indicating that the biochar has reached equilibrium in adsorption of Pb(II) in water. It can also be seen from the figure that at an adsorption time of 3 h, the adsorption capacities of B450, B550, and B650 biochars for Pb(II) are 5.738 mg/g, 5.85 mg/g, and 6.138 mg/g, respectively.

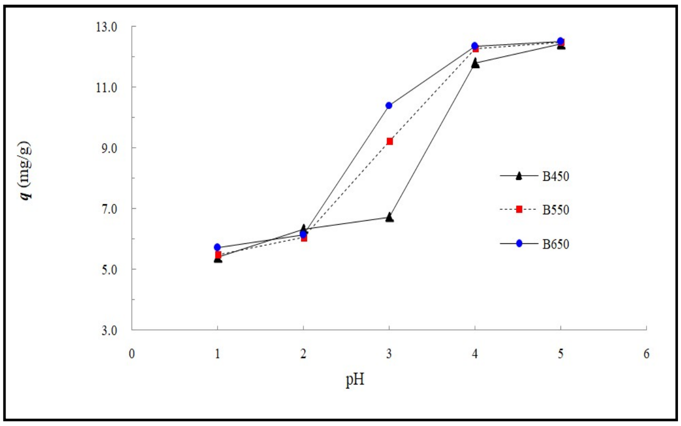

Figure 4. Effect of different pH values on the adsorption of Pb(II)

Therefore, when other conditions are constant, the best adsorption effect of biochar under B650 for Pb(II) in water is achieved when the adsorption time reaches 3 h.

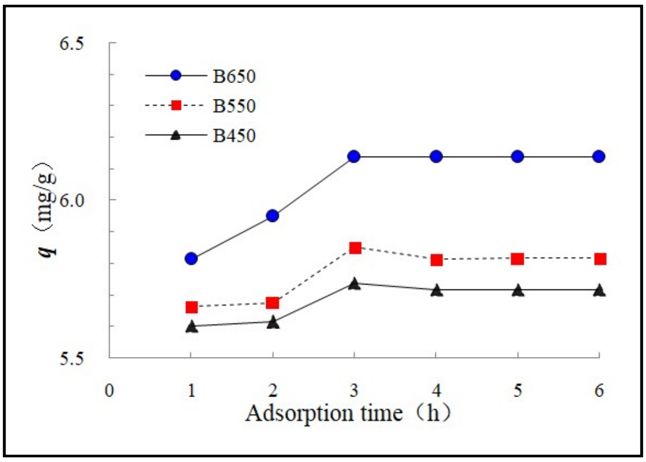

Figure 5. Effect of adsorption time on the adsorption of Pb(II)

CONCLUSION

The three types of biochar prepared from coconut shells at different temperatures have good adsorption effects on Pb(II) in water. When the temperature is 30 °C, the initial solution concentration is 100 mg/L, the addition amount is 8 g/L, the pH is 5, and the adsorption time is 3 h, the adsorption effect of biochar B650 on Pb(II) in water is the best. The isothermal adsorption curves of Pb(II) in water by the three biochars can be well fitted by Freundlich and Langmuir equations, among which B650 has the strongest adsorption capacity. This study can provide a theoretical basis and technical reference for the utilization of agricultural and forestry wastes for the treatment of heavy metals in wastewater.

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