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Sustainable utilization of agricultural waste: Adsorption performance of magnetic polyethyleneimine-modified peanut shell biochar for eco-friendly Congo red removal in dyeing wastewater

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Abstract: Agricultural wastes are various types of residual materials generated in the process of agricultural production, such as crop stalks, fruit shells, etc., which are so large in quantity that they not only cause waste of resources but also may lead to many problems, such as environmental pollution, if they are not handled properly. This study focuses on the sustainable utilization of agricultural waste - peanut shells, especially the modification of peanut shell biomass charcoal and its application in dye wastewater treatment, aiming to achieve the removal of Congo red dye from wastewater in an eco-friendly manner. The study evaluated the effect of Fe3O4-PEI-BC on CR adsorption by varying the reaction temperature, reaction time, adsorbent dosage, pH, and initial dye concentration. The experimental results showed that the adsorption efficiency was higher as the temperature was increased to 35 °C, the reaction time was prolonged, the concentration of adsorbent was more than 40 mg/150 mg CR/L, the pH value was 2, and the initial concentration of CR was 50 mg/L. This study not only provides a new idea for the resource utilization of agricultural waste but also provides an effective technical solution for the environmental protection treatment of printing and dyeing wastewater.
Keywords: *Agricultural waste, Adsorption performance, Biochar, Congo red, Green materials, Sustainable utilization.*

1. Introduction

China is a large agricultural country, and the amount of agricultural waste generated is huge. According to statistics, the annual output of crop straw and livestock manure, which are the main agricultural wastes in China, is 0.9 billion tons (in terms of dry matter) and 3.8 billion tons, respectively [1]. With the increase in the level of agricultural production in China, the generation of agricultural waste has increased at an average annual rate of 5-10%. China has the largest annual peanut production in the world, and peanut shells, as an adjunct to peanuts, account for about 30% of the total peanut production, i.e., about 5.25 million tons per year [2]. However, despite the abundant resources of peanut shells, their utilization rate is not high, which not only causes pollution of the environment, but also leads to a serious waste of resources [3]. Therefore, this paper focuses on exploring the resource utilization of peanut shells as agricultural wastes.

At present, fertilizer, feed and fuel of agricultural waste is still the main development direction of treatment. However, in today's market economy and industrialized operation, it is no longer adapted to the needs of development [4]. Aiming at high-value product development, comprehensive utilization of agricultural waste resources is one of its development trends. Among them, biomass industry is the whole process of transforming biomass raw materials such as energy plants and agricultural wastes into high value-added biomass energy, biomaterials, petroleum product substitutes and by-products, and other environmentally friendly products through industrialization using chemical or biological technologies. In recent years, biochar prepared from biomass has attracted much attention as a green

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renewable energy source in the field of wastewater treatment [5]. Not only does it help to reduce the accumulation of agricultural waste and solve the challenge of agricultural waste disposal, but it also solves the challenge of compliant wastewater discharge.

However, biochar is still deficient in adsorption and stability. In order to further improve the performance of biochar for wastewater treatment, physical or chemical methods are usually used to modify biochar to increase its surface area and improve its microporous structure. Currently, researchers have carried out a lot of studies on the effect of biochar on wastewater treatment under the modification methods of mechanical milling, microwave radiation, plasma treatment, acid and alkali treatment, potassium permanganate oxidation and metal oxide loading. But there is less research on the modification effect of magnetic polyethyleneimine (Fe_3O_4-PEI) biochar, which is simple and inexpensive to prepare, and can improve the adsorption capacity of anionic dyes, and can be utilized to recycle the magnetic properties in a simple and low-cost process [6].

Therefore, in this study, CR, an anionic dye which is widely present and difficult to treat, was selected as the target pollutant, and the adsorption effect of magnetic polyethyleneimine-modified peanut shells-biochar (Fe_3O_4-PEI-BC) on CR was analyzed at different temperatures, response times, pH, adsorbent dosages, and initial concentrations of CR. The aim of the study was to confirm the optimal adsorption conditions and providing scientific guidance and theoretical basis for the resource utilization of agricultural wastes.

2. Literature Review

The sustainable use of agricultural waste is an important issue for global environmental management and resource optimization. As agricultural production increases, so do the environmental consequences associated with waste generation. Agricultural wastes, including crop residues and livestock manure, account for a significant portion of global waste production and, if not handled properly, will pollute the environment and pose health risks. However, with the progress of science and technology and the enhancement of environmental awareness, the resource utilization of agricultural waste has gradually become a research hotspot. Among them, peanut shells, as a common agricultural waste, is rich in cellulose and lignin structure, which provides a possibility for its conversion into high-performance biochar.

BC is a kind of green and sustainable material with porous, high surface area, high aromatization and relatively stable physicochemical properties, which is obtained by cracking carbon-rich biomass under anaerobic or anoxic conditions at high temperatures(<700℃) [7]. These unique physicochemical characteristics of BC have enabled its wide application in many fields such as energy recovery, carbon sequestration, soil remediation, and wastewater treatment [8, 9].

The National Development and Reform Commission , the Ministry of Housing and Urban-Rural Development , and the Ministry of Ecology and Environment jointly issued the "*Implementing Opinions on Promoting Synergistic Efficiency of Wastewater Treatment in Reducing Pollution, Reducing Carbon, and Increasing Efficiency*" National Development and Reform Commission [10] which emphasizes the importance of the environmentally sound treatment of wastewater and the recovery and utilization of resources, in order to promote the formation of a green and cyclical development model. China's annual printing and dyeing wastewater emissions are about 2 to 2.3 billion tons per year, accounting for about 11% of the national wastewater emissions [11, 12]. Although the emissions and the proportion are not high, its organic content is high, its composition is complex, and some of it contains carcinogenic and teratogenic organic substances or heavy metals. In order to cope with the environmental problems caused by the discharge of printing and dyeing wastewater and actively respond to the government's new policy of green development, BC has attracted great attention as a sustainable and ecological development of sewage adsorbent.

When biochar (BC) is applied directly as an adsorbent, its effectiveness in wastewater treatment is relatively limited. Currently, in-depth studies on the efficacy of biochar in wastewater treatment under a variety of modification methods have been widely conducted, which cover mechanical milling,

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microwave radiation, plasma treatment, acid and alkali treatment, potassium permanganate oxidation, and metal oxide loading, etc.Xue, et al. [13] found that hydrogen peroxide modification enhanced the adsorption of waterborne heavy metals by biochar derived from hydrothermal carbonization of peanut shells capacity with good results. Ahmed, et al. [14] found that modified biochar derived from watermelon seeds enhanced the adsorption capacity of lead (II) ions in solution to 60.87 mg/g.

Among them, PEI is widely used in the surface modification of biochar because of its low price, environmental protection and renewable characteristics. The amino group (-NH2) in polyethyleneimine reacts chemically with the functional groups on the surface of activated charcoal (e.g., hydroxyl, carboxyl, etc.) and forms covalent or ionic bonds, which increases the hydrophilicity and polarity of the surface of activated charcoal, and makes it more likely to interact with polar substances (e.g., water, organic matter, etc.). During the modification process, polyethyleneimine may block some of the micropores of the activated carbon, but at the same time, it will also introduce new functional groups and pore structures, which will make the pore structure of the activated carbon more complex and diversified. This significantly improves the adsorption capacity of activated carbon for pollutants such as heavy metals, organics and phosphates Tian [15]. Zhang, et al. [16] prepared PEI-modified activated carbon (AC) to obtain PEI-AC by impregnation with activated carbon and used it for the adsorption of gaseous formaldehyde. The results showed that the adsorption capacity of PEI-AC for formaldehyde was 1.67 times that of the unmodified activated carbon. Gao [17] used PEI to modify AC and applied it to the adsorption of Cd (II) in wastewater. The results indicated that the adsorption of Cd (II) by PEI/AC mainly occurred on the —NH and $—NH_2$ groups on the surface of PEI/AC, with an adsorption capacity reaching 45 mg/g, which was 35 mg/g higher than that of the unmodified activated carbon. It can be seen that using PEI to modify activated carbon can effectively adsorb pollutants in water, and it has broad potential in wastewater treatment [18]. Other scholars have also studied the adsorption effect of modified biochar. For example, Xue, et al. [13] confirmed that the modification with hydrogen peroxide enhanced the ability of biochar derived from the hydrothermal carbonization of peanut shells to adsorb heavy metals in water, achieving good results. Ahmed, et al. [14] found that the modified biochar extracted from watermelon seeds improved the adsorption capacity of lead (II) ions in the solution, with an adsorption capacity reaching 60.87 mg/g.

Magnetic biochar (MBC) is a type of biochar with magnetic properties, which is obtained by introducing magnetic precursor substances (such as iron salt solutions, natural iron minerals, and iron oxides) on the basis of biomass or BC [19, 20]. MBC can efficiently remove heavy metals from water through surface adsorption, ion exchange and complexation, and also adsorb organic matter containing π-π interactions, hydrogen bonding, and hydrophobic interactions to purify the wastewater [21, 22]. Meanwhile, the purified water can be easily separated from solid-liquid by magnetic apparatus, which in turn recovers MBC in the solid phase Yi, et al. [23]. Zhu, et al. [24] used cow dung as a biomass raw material to prepare MBC at pyrolysis temperatures of 200, 400, and 600 ℃, and it was found that, with the increase of pyrolysis temperature, the specific surface area of MBC increased from 4.71, 18.52 to 188.56 m^2 /g, showing a gradual increasing trend. This may be due to the fact that the iron oxides of MBC existed as γ-Fe_2O_3 at low temperatures, and the iron oxides existed in the form of Fe_3O_4 when exceeding 400 ℃. Li, et al. [25] found that when the pyrolysis temperature reached 800 ℃, the presence of iron oxides blocked the development of pores, which led to the phenomenon of decreasing the specific surface area of MBC. Therefore, the specific surface area of MBC showed a tendency of increasing and then decreasing with the increase of pyrolysis temperature.

In addition, research findings indicate that the adsorption capacity of MBC is closely related to the types of biomass, biomass type directly affects the nature of MBC. For example, Trakal, et al. [26] selected five different types of biomass (nut shells, wheat straws, grapevines, grape skins, and plum pits) to prepare the corresponding BC and MBC. It was found that the cation exchange capacity (CEC) of magnetically modified nut shells and plum shells magnetic biochar increased by 3.3 and 1.5 times, respectively, compared with the original biochar, and their ability to adsorb Pb and Cd was also significantly enhanced (10-16 and 5-7 times, respectively). In contrast, the changes in the CEC values of the MBC from wheat straws, grapevines, and grape skins after magnetic modification were very small (less than 1 time), and correspondingly, the changes in their abilities to adsorb Pb and Cd were also very small (not exceeding 1 time).

In summary, facing the concept of sustainable development, biochar can be used as a green material for sewage treatment. However, the adsorption performance of biochar itself is not strong and recovery is difficult, so the activated carbon can be modified so as to improve the adsorption effect of activated carbon on organic matter and heavy metals in wastewater, etc. and to improve the capacity of wastewater treatment. PEI, as a common and effective biochar charcoal modifier, adsorbs pollutants in the water efficiently, and it has a wide range of potentials for the treatment of wastewater. The introduction of magnetic precursors to biochar not only further improves the adsorption capacity of biochar, but also enables efficient recycling and reuse through magnetic instruments.The influencing factors of the adsorption capacity of MBC are mainly temperature and type of biomass.

In this paper, two modification methods, namely PEI and MBC, are innovatively combined. Using peanut shells as biomass raw materials, the modified biochar Fe_3O_4-PEI-BC is prepared. The influences of Fe_3O_4-PEI-BC on the chromaticity of CR dye solution under different temperatures, response times, adsorbent dosages, pH values and initial concentrations of CR are systematically studied. The aim is to obtain the optimal reaction conditions and provide a scientific basis and theoretical reference for the application of biochar in the treatment of printing and dyeing wastewater.

3. Materials and Methods

3.1. Experimental Materials

Peanut shells were purchased online and originated from Shandong, China. Shandong is a major peanut-producing area, which can ensure the quantity and quality of the purchased peanut shells. PEI was purchased from Shanghai HanSi Chemical Co., Ltd. In addition, glutaraldehyde, absolute ethanol, Fe_3O_4, and NaOH used in the study were respectively purchased from Chengdu Jinshan Chemical Reagent Co., Ltd., Baiyin Liangyou Chemical Products Co., Ltd., Shanghai Shanhai Gongxuetuan Experimental No. 2 Factory, and Xilong Technology Co., Ltd. The manufacturers of potassium carbonate (98%) and dilute hydrochloric acid (37%) are Guangdong Wengjiang Chemical Reagent Co., Ltd. The manufacturer of CR is Beijing Chemical Works. All reagents are of analytical purity, and the experimental water used is deionized water.

3.2. Preparation of Fe_3O_4-PEI-BC

3.2.1. Preparation of Biochar

Place 10 grams of pretreated peanut shell powder into a 150-milliliter crucible, and then send the crucible into a tube furnace. Under the protective atmosphere of nitrogen (N_2), carry out the carbonization treatment at a heating rate of 10 °C/minute by adjusting the heating condition parameters (ranging from 600 °C to 1000 °C). After it cools down to room temperature naturally, take out the obtained product and soak it in a 1 mol/L dilute hydrochloric acid solution for 24 hours to remove the ash content. After soaking, wash the sample with pure water several times until its pH value reaches the range of 6 - 7. Then, place the washed sample in an oven at 110 °C for drying for 4 hours. After drying, grind the sample with a mortar and sieve it through a 100-mesh sieve. Finally, store the sieved sample in a dry container to prevent it from absorbing moisture from the air [20]. The obtained sample material is labeled as BC for subsequent experiments and analyses.

3.2.2. Preparation of PEI-Modified Biochar

First, accurately weigh 3.0 g of PEI and dissolve it in 150 mL of absolute ethanol. Next, transfer this solution to an Erlenmeyer flask containing 9.0 g of peanut shells and stir the mixture for 24 hours. After that, add the treated peanut shells to 300 mL of a solution containing 1% glutaraldehyde and continue to stir for 1 hour. Subsequently, discard the clear liquid on the upper layer and wash the peanut shells with distilled water and absolute ethanol respectively until the clear liquid on the upper layer becomes neutral. Finally, place the washed peanut shells in an oven at 60 °C for drying, and the resulting product is the PEI-modified biochar (PEI-BC).

3.2.3. Preparation of Magnetically Modified Biochar

Mix Fe_3O_4 and PEI-BC in a ratio of 1:2 and add them into a beaker. Then, add ultrapure water to fix the volume to 100 mL and continuously stir the mixture at 60 °C for 24 hours. During the stirring process, Fe_3O_4 will evenly adhere to the surface of PEI-BC or penetrate into its pore structure to combine with PEI-BC, ultimately forming the magnetic PEI-modified peanut shell biochar (i.e., Fe_3O_4-PEI-BC). After that, centrifuge the above mixture and dry the solid substance obtained by centrifugation at 75 °C, and finally, Fe_3O_4-PEI-BC is prepared.

3.3 Adsorption Experiment

3.3.1. Preparation of Dye Solution

Accurately weigh 1 g of CR dye in a beaker. Then, add an appropriate amount of deionized water and stir thoroughly to ensure complete dissolution. Next, transfer the solution to a 1000 mL volumetric flask and continue to add deionized water to the volumetric flask until it reaches the calibration mark, thus preparing a 1 g/L standard solution of CR dye. The CR solutions required in subsequent experiments are all diluted from this standard solution as needed.

3.3.2. Adsorption Performance Test

3.3.2.1. Effect of the Reaction Temperature

Pour 100 mL of the CR solution with a concentration of 150 mg/L into a 250 mL conical flask. Then, add 10 mg of the Fe_3O_4-PEI-BC adsorbent into the flask. Conduct measurements at temperatures of 15 °C, 25 °C, and 35 °C respectively, record the adsorption values, and then calculate the adsorption removal rate.

3.3.2.2. Effect of Adsorption Time

Pour 100 mL of the CR solution with a concentration of 150 mg/L respectively into several 250 mL conical flasks, and then add 10 mg of the Fe_3O_4-PEI-BC adsorbent to each flask. After that, subject these mixtures to oscillation for 2 hours, 4 hours, 6 hours, 8 hours, 10 hours, and 12 hours respectively. Upon completion of the oscillation, take the supernatant liquid and centrifuge it at 30 °C. Then measure its absorbance and calculate the adsorption removal rate based on it.

3.3.2.3. Effect of Adsorption Dosage

Pour 100 mL of the 150 - mg/L CR solution into a 250 - mL conical flask. Then, add 2.5 mg, 5 mg, 10 mg, 15 mg, 20 mg and 30 mg of Fe_3O_4 - PEI - BC adsorbent respectively. After that, measure its absorbance and calculate the corresponding adsorption removal rate.

3.3.2.4. Effect of pH

Pour 100 mL of the 150 - mg/L CR solution into a 250 - mL conical flask. Then, add 10 mg of the Fe_3O_4 - PEI - BC adsorbent. Adjust the pH value to a range between 1 and 12. Oscillate the mixture at 30 °C for 2 hours, and then extract the supernatant for centrifugation. By measuring the absorbance, the adsorption efficiency and the pollutant removal rate can be calculated.

3.3.2.5. Effect of initial concentration of CR

Prepare CR solutions with different concentrations ranging from 50 mg/L to 300 mg/L. Take 100 mL of the CR solutions with different concentrations and place them respectively in 250 mL conical flasks. Subsequently, add 10 mg of the magnetic-PEI-biochar adsorbent. Calculate the adsorption removal efficiency of CR in the solutions by measuring the absorbance.

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4. Results and Discussion

4.1. Effect of Reaction Temperature

As shown in Figure 1, with the temperature increasing from 15 °C to 35 °C, the adsorption efficiency of Fe_3O_4-PEI-BC for CR exhibits a significant upward trend, rising from 17.6% to 23.4%. Meanwhile, its adsorption capacity also experiences a remarkable increase from 264 mg g^{-1} to 351 mg g^{-1}. This change can be attributed to two major factors. Firstly, as the temperature rises, the solubility of CR in water correspondingly increases, resulting in more CR molecules being in a free state, which in turn enhances the possibility of them being captured by the adsorbent. Secondly, the increase in temperature may also promote the interaction between CR molecules and the Fe_3O_4-PEI-BC adsorbent. Under high-temperature conditions, the thermal motion among molecules intensifies, causing both the collision frequency and collision intensity between CR molecules and the active sites on the adsorbent surface to increase, thereby strengthening the binding force between them and further improving the adsorption efficiency. This conclusion is consistent with the research findings of Wekoye, et al. [27].

In addition, it is worth noting that although an increase in temperature usually intensifies the thermal motion and collision frequency of molecules, which is beneficial to the adsorption process, an excessively high temperature may also lead to the instability of the adsorbent structure, thereby affecting the further improvement of the adsorption efficiency and adsorption capacity. For example, Khan, et al. [28] found in their study on the removal of CR dye using the anion exchange membrane EBTAC that as the temperature gradually increased (from 19 °C to 50 °C), the adsorption efficiency of CR showed a slight downward trend. This result implies that a potential exothermic process may be involved when using the anion exchange EBTAC membrane for CR adsorption.

Although in general, an increase in temperature will intensify the thermal motion of molecules and increase the collision frequency between molecules, which is theoretically beneficial to the progress of the adsorption reaction, an excessively high temperature may also have adverse effects. On the one hand, high temperature may cause the instability of the internal structure of the adsorbent, affecting its adsorption performance; on the other hand, high temperature may also prompt the target molecule (such as CR) to undergo degradation or structural changes, thereby reducing its ability to be effectively captured by the adsorbent. Therefore, in practical applications, it is necessary to comprehensively consider the dual effects of temperature on the adsorption process in order to find the optimal adsorption temperature and achieve the goal of efficient and economical wastewater treatment.

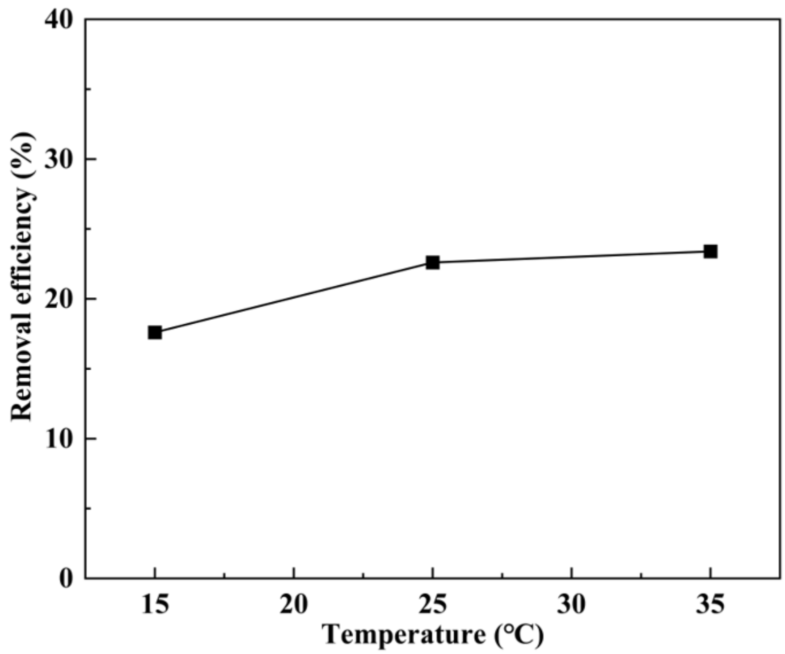

Figure 1.
Effect of temperature on the removal efficiency of CR by Fe_3O_4-PEI-BC.

4.2. Effect of Reaction Time

The adsorption rate at different response times and the subsequent adsorption time to reach adsorption equilibrium are important parameters in the adsorption process [29]. According to Figure 2, in the initial stage of adsorption, as the reaction time gradually extends, the removal efficiency of CR shows a rapid growth trend. This significant growth trend is mainly attributed to the large number of unoccupied adsorption active sites existing on the surface of Fe_3O_4-PEI-BC. These sites have relatively high surface energy and affinity, and they can efficiently capture and fix the CR molecules in the solution, thus realizing the rapid rise of the CR removal rate. However, as the reaction continues, the available adsorption sites on the surface of Fe_3O_4-PEI-BC are gradually occupied by CR molecules, resulting in a significant reduction in the number of remaining unoccupied sites, and further causing the growth rate of the adsorption efficiency to gradually slow down. At this time, the removal rate of CR stabilizes at 21.1%, and the adsorption equilibrium amount reaches 316.5 mg g^{-1}.

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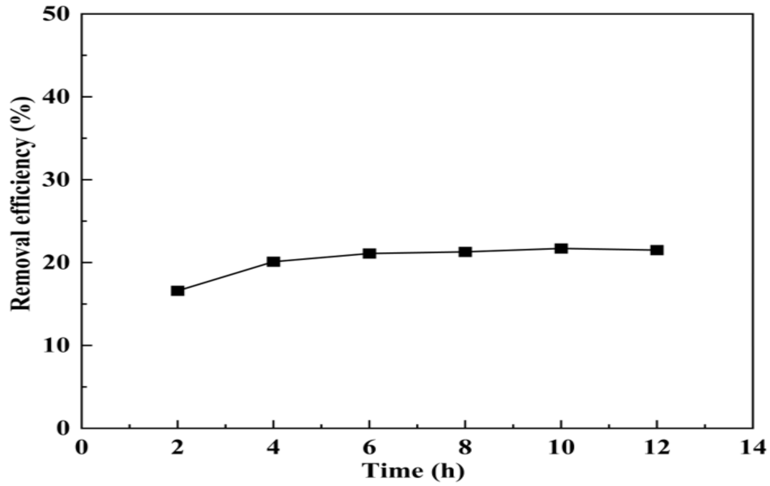

Figure 2.
Effect of response time on the removal efficiency of CR by Fe_3O_4-PEI-BC.

4.3. Effect of Adsorbent Amount

Figure 3 depicts in detail the specific impact of the dosage of the Fe_3O_4-PEI-BC adsorbent on the removal rate of CR. As can be seen from the figure, the removal rate of the CR dye fluctuates within the range of 28.9% to 96.7%. Specifically, with the gradual increase in the dosage of the Fe_3O_4-PEI-BC adsorbent, the removal rate of CR also shows an increasing trend. It is particularly noteworthy that during the process of increasing the adsorbent dosage from 2.5 mg to 10 mg, the removal rate jumps sharply from 28.9% to 94.3%. This significant change can be attributed to the fact that, under the premise of keeping other experimental conditions constant, increasing the dosage of Fe_3O_4-PEI-BC essentially provides a broader adsorption surface area and more abundant adsorption sites for the adsorption process. These newly added surface areas and sites are like "traps" that can capture and fix the CR molecules in the solution more efficiently, thereby significantly enhancing the removal efficiency of CR under the same concentration conditions. This finding is consistent with the research results of Wang, et al. [8].

However, when the dosage of the Fe_3O_4-PEI-BC adsorbent exceeds the threshold of 10 mg, the growth rate of the removal rate begins to show a trend of slowing down and gradually approaches a relatively stable equilibrium state. This phenomenon can be explained from the perspective of the interaction between adsorbent particles. At high dosages, due to the shortened spatial distance between adsorbent particles, they are more likely to aggregate with each other. This aggregation phenomenon may cause some of the originally available adsorption sites to be masked by other particles or not be fully utilized due to steric hindrance effects. As a result, the number of active adsorption sites accessible to the CR dye molecules will correspondingly decrease [30]. Therefore, although the dosage of the adsorbent is continuously increased, the improvement in the removal rate becomes less significant. This phenomenon reveals that the relationship between the adsorbent dosage and the removal efficiency is not a simple linear one, but there exists an optimal dosage range.

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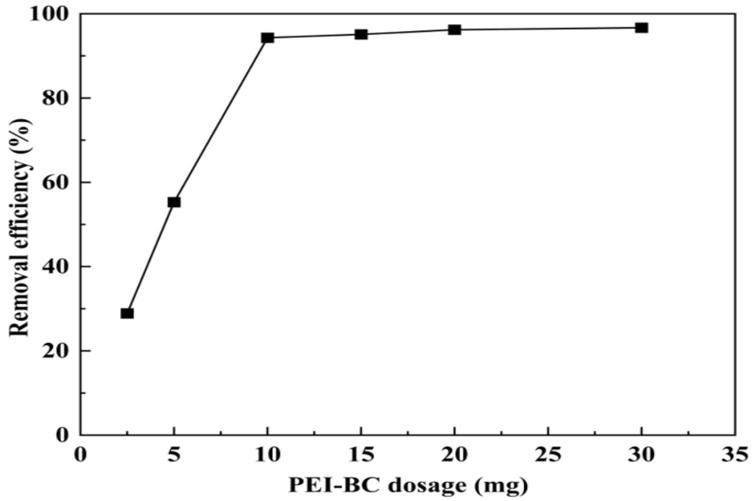

Figure 3.
Effect of Fe_3O_4-PEI-BC dosage on CR removal efficiency.

4.4. Effect of pH

The pH value of the solution plays an important role in the process of the Fe_3O_4-PEI-BC adsorbent removing CR dye molecules. It profoundly affects the charge properties on the surface of the adsorbent as well as the existing forms of CR molecules, and thus has a crucial impact on the overall adsorption performance. Through in-depth analysis of the data provided in Figure 4, it can be found that under acidic conditions, the adsorption efficiency of Fe_3O_4-PEI-BC for CR is the highest. This phenomenon can be attributed to the strong electrostatic attraction between the negatively charged CR dye molecules and the positive charges on the surface of the biochar impregnated with PEI. This interaction greatly promotes the attachment and fixation of CR molecules on the surface of the adsorbent [31]. Specifically, under the extremely acidic condition with a pH value of 2, the adsorption efficiency of Fe_3O_4-PEI-BC for CR can be as high as 21.2%, fully demonstrating its superior adsorption performance in an acidic environment.

However, as the pH value of the solution gradually increases, the removal rate of CR begins to show an obvious downward trend. Especially in the neutral to weakly alkaline range where the pH value is between 4 and 7, the decline in the removal rate is particularly significant. The reason behind this change is closely related to the change in the charge properties on the surface of the adsorbent. As the pH value increases, the charge state on the surface of the adsorbent gradually changes from positive to negative. When the pH value of the solution exceeds the point of zero charge (pZC) of the adsorbent, the surface of the adsorbent will carry the same negative charge as the CR molecules, which leads to a strong repulsive force between them, thus greatly reducing the adsorption efficiency.

When the pH value of the solution is further increased to 10, the adsorption efficiency has dropped to 0.9%. This is because under such a high pH value condition, the surface of the adsorbent is almost completely negatively charged, forming a strong repulsive force field with the CR molecules that are also negatively charged, making it almost impossible to carry out effective adsorption [32]. Therefore, we can conclude that the pH value of the solution is one of the key factors affecting the efficiency of the Fe_3O_4-PEI-BC adsorbent in removing CR dye molecules. By precisely controlling the pH value of the solution, we can further optimize the adsorption process and improve the removal efficiency.

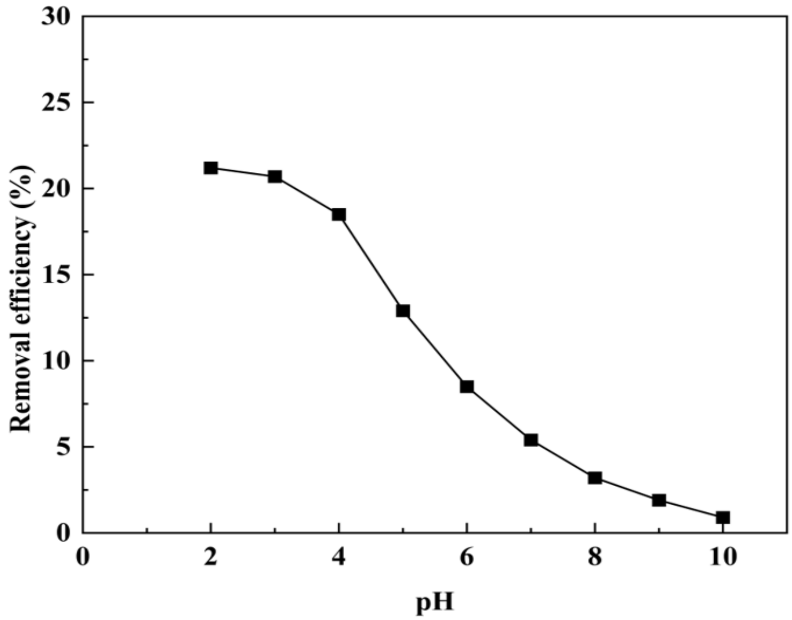

Figure 4.
Effect of reaction time on the removal efficiency of CR by Fe_3O_4-PEI-BC.

4.5. Effect of Initial Concentration of CR

On the premise of keeping other experimental conditions constant, this study thoroughly investigated the impact of the initial concentration variable on the performance of Fe_3O_4-PEI-BC in adsorbing CR dye. According to Figure 5, as the initial CR concentration gradually increases, the CR adsorption efficiency of an equal amount of Fe_3O_4-PEI-BC adsorbent shows a significant downward trend. Specifically, when the initial CR concentration is 50 mg/L, the adsorption efficiency of Fe_3O_4-PEI-BC is as high as 54.6%. However, as the initial CR concentration climbs to the highest level of 300 mg/L, the adsorption efficiency drops sharply to only 10.6%.

By delving into the reasons behind this phenomenon, we find that for a given amount of Fe_3O_4-PEI-BC adsorbent, the number of active sites on its surface available for CR molecules to bind is limited. When the CR concentration is low, these active sites can be fully exposed and have effective physical or chemical interactions with CR molecules, thus achieving a high adsorption efficiency. However, as the CR concentration gradually increases, more and more CR molecules start to compete for these limited adsorption sites, causing some CR molecules to fail to be effectively adsorbed due to their inability to find suitable binding positions, and thus resulting in a gradual decline in the overall removal rate. When the adsorption sites reach a saturated state, that is, when all the available sites have been occupied by CR molecules, even if the CR concentration is continuously increased, the adsorption efficiency cannot be further improved, because there are no additional active sites available for use on the surface of the adsorbent [33].

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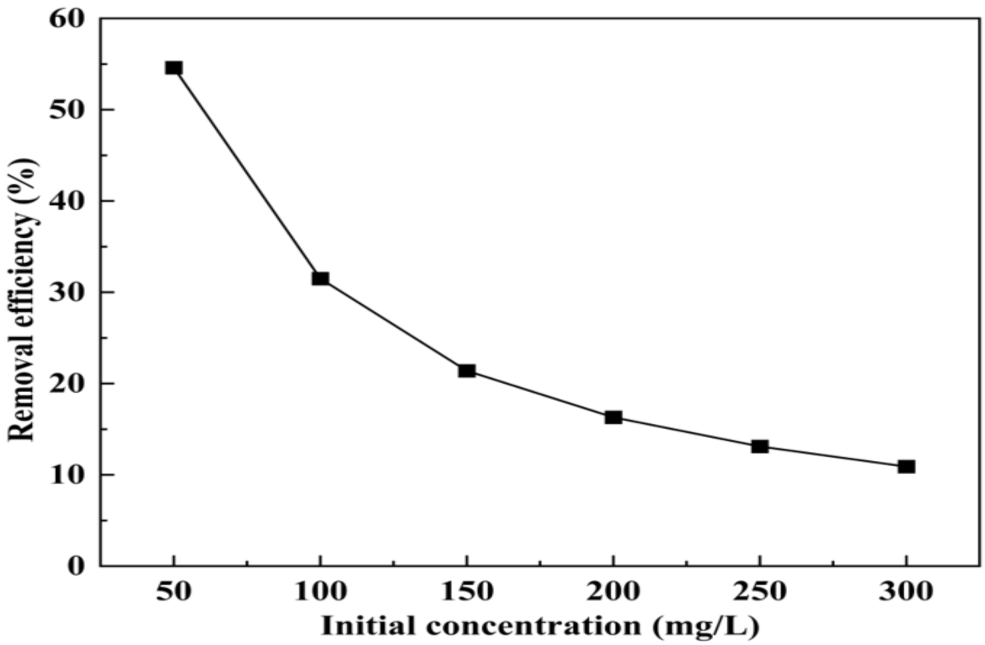

Figure 5.
Effect of different initial concentrations of CR on the removal efficiency of CR by Fe3O4-PEI-BC.

5. Conclusion

In summary, in this study, Fe_3O_4-PEI-BC biochar material was successfully prepared from peanut shells, an agricultural waste, and its adsorption performance on CR in printing and dyeing wastewater was systematically studied. The results show that Fe_3O_4-PEI-BC has a good adsorption effect on CR, and the adsorption process is affected by multiple factors, including reaction temperature, adsorption time, adsorption dosage, pH value, and the initial concentration of CR.

Specifically, as the temperature rises, both the adsorption efficiency and adsorption capacity of CR increase. This may be related to the increased solubility of CR molecules in water and the intensified thermal motion among molecules. The impact of response time on the adsorption efficiency indicates that in the initial stage, the adsorption efficiency increases rapidly. However, as the reaction time extends, the growth rate of the adsorption efficiency slows down and reaches the adsorption equilibrium state after 6 hours. The increase in the adsorption dosage significantly improves the removal rate of CR, especially within the range from 2.5 mg to 10 mg. However, when the adsorption dosage exceeds 10 mg, the growth rate of the removal rate begins to slow down, suggesting that there is an optimal adsorption dosage. The pH value of the solution has a decisive impact on the adsorption efficiency. Under acidic conditions, due to the effect of electrostatic attraction, the adsorption efficiency of Fe_3O_4-PEI-BC for CR is the highest. As the pH value rises, the adsorption efficiency drops rapidly. Especially when the pH value exceeds the point of zero charge of the adsorbent, the adsorption efficiency is significantly reduced. The initial concentration of CR has a negative impact on the adsorption efficiency. As the initial concentration of CR increases, the adsorption efficiency of the same dosage of Fe_3O_4-PEI-BC for CR gradually decreases. This may be because the limited adsorption sites cannot meet the competitive demands of high-concentration CR molecules.

Based on the above findings, Fe_3O_4-PEI-BC, as a new type of magnetically modified biochar, not only effectively removes dye pollutants from wastewater, but also realizes the reuse of agricultural wastes. In the future, the energy utilization of agricultural waste is the main strategic orientation.

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Resource utilization of agricultural waste will play an important role in the circular economy and make a significant contribution to the sustainable development of agriculture and energy security in China.

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Transparency:
The authors confirm that the manuscript is an honest, accurate and transparent account of the study that no vital features of the study have been omitted and that any discrepancies from the study as planned have been explained. This study followed all ethical practices during writing.

Competing Interests:
The authors declare that they have no competing interests.

Authors' Contributions:
LS methodology, software, formal analysis, writing original draft,visualization. NN and AAB conceptualization, validation, writing - reviewand editing, supervision. All authors have read and agreed to the published version of the manuscript.

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