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Experimental Study on the Direct Removal of Cadmium from Aqueous Solution Using Peanut-Activated Biochar without Residual Rinse

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Abstract

The use of agricultural waste to produce low-cost activated carbon (AC) adsorbents for cadmium ion (Cd2+) removal in sewage has drawn more attentions. Conventionally, for biochar activated by alkaloids, pH adjustment is required, and it involves operations, such as washing and sieving. Given that wastewater containing Cd^{2+} is typically acidic, the adsorption experiment of Cd²⁺ must be conducted immediately after the preparation of AC, which may lead to crucial improvements in the current treatment process for Cd²⁺-containing sewage. To explore the direct application process of biochar prepared through alkaloid-impregnated activation for the removal of Cd²⁺ in water, this study used peanut shells as a raw material (RM) and potassium hydroxide (KOH) as a modifier/activator to prepare biochar. After subjecting the peanut shells to activation with low-concentration KOH and pyrolysis at 600 °C, proximate analysis and surface morphology comparison of the obtained biochar were performed. Meanwhile, the untreated biochar and 10% KOH-AC were used in adsorption experiments, with a scanning electron microscope utilized for the surface study. The results show the following: (1) Sieving can effectively eliminate ash from AC but has limited improvement on the removal rate of Cd²⁺ (Cd-RR) and cause mass loss, which reduces the comprehensive removal efficiency. (2) The 10% KOH-activated biochar at 3.0 and 1.0 g/L achieves the best Cd-RR of 99.15% and 98.99%, respectively. The crushed KOH-impregnated peanut shell AC can remediate Cd²⁺ wastewater directly and has advantages in terms of RM cost, preparation process, and removal benefits compared with traditional ones. The conclusions provide a novel method for the treatment of Cd2+containing sewage using peanut shell AC.

Keywords: Peanut shells, KOH activation, Activated carbon, Cd2+, Removal rate

1. Introduction

Cadmium (Cd), a heavy metal, can be dispersed in soil solutions or surface water, and it exerts toxic effects on humans and diverse living organisms [1, 2]. In 2014, China's environmental authorities revealed that the percentage of Cd exceeding the standard in heavy metalcontaminated soils across China reached 7% [3]. In Dingshu town, which is located in the west of Taihu Lake, an indepth analysis suggested that the overall amount of Cd discharged into the environment in 2015 was around 43.5 kg, with approximately 90.4% discharged into water bodies. The national discharge of heavy metals from water pollution was 182.54 tons, and all were observed in the seven major river basins [4]. Once Cd enters water, it can permeate into daily life and production via pipelines or irrigation systems, which renders its collection and treatment procedures extremely difficult. Considering that water heavy-metal pollution represents a considerable environmental burden and Cd ranks among the highly toxic heavy metals that exist in wastewater [5], treatment before its spread is highly important.

Under acidic circumstances, cadmium ion (Cd^{2+}) displays a high reactivity [6-8]. Cd can persist in solutions in

the form of soluble complexes with anions, such as CdCl⁺ and $Cd(SO_4)_2^{2^-}$, along with dissolved organic substances. Meanwhile, adsorption and precipitation will diminish the concentration of the majority of other heavy metals [9]. Among these influencing factors, the concentration of hydroxide ions (OH⁻) exerts a substantial effect on the morphological distribution pattern of Cd [10, 11].

Biochar is notable for its remarkable chemical and biological stability. This substance is endowed with a porous structure and encompasses an extensive specific surface area. Its surface harbors a great quantity of oxygen-containing functional groups, such as carboxyl (-COOH), hydroxyl (-OH), and carbonyl (-CO). These functional groups can furnish adsorption sites for heavy-metal ions and organic contaminants, which facilitates the formation of stable complexes and augments the removal efficiency of heavy metals [12]. The most promising and prospective application approach lies in the utilization of agricultural residues in the fabrication of low-cost biochar adsorbents to efficiently remove Cd²⁺ from wastewater [13-17]. This strategy not only offers a sustainable solution by recycling agricultural by-products but also holds a great potential in addressing the pressing issue of heavy-metal pollution in aqueous environments. Moreover, it plays a crucial role in the

development of environmentally friendly and economically viable wastewater treatment technologies.

Peanut shells are rich in lignin and fiber constituents [18], which possess diverse functional groups, such as -OH and -COOH, that confer peanut shells with a remarkable adsorption capacity [19]. These properties make peanut shells a potentially valuable material in various adsorptionrelated applications and environmental remediation processes. The chemically activated carbon (AC) obtained from peanut shells demonstrates a high level of efficacy in Cd adsorption. According to the statistical analysis presented by the National Bureau of Statistics, the projected peanut production in China in 2023 was approximately 19.2307 million tons [20]. Empirical studies have demonstrated that each variety of AC manifests distinct behaviors in the presence of different metal ions [21]. Cheng et al. conducted a comprehensive study on 22 types of materials and identified peanut shell biochar as the most potent material for the remediation of Cd in aqueous phase among these 22 candidates [22]. Typically, the adsorption equilibrium is attained within approximately 12 h. Under the optimized process conditions, the removal rate can reach as high as 99.9%. In addition, the pyrolysis temperature corresponding to the optimal adsorption performance of biochar is 600 °C [23]. Peanut shell-based biochar has an outstanding pore structure, enhanced surface cleanliness, and facile adsorption properties. At the pyrolysis temperature, the quantity and density of oxygen-containing functional groups on the biochar surface can be maintained, and the decomposition of its organic constituents can be avoided [24, 25].

Chemical activation is more efficient and results in a larger surface area compared with physical activation [26]. Two chemical activation methods are primarily used for biochar. The first involves the impregnation of the carbon precursor with a chemical activator followed by pyrolysis. The second entails an initial carbonization, subsequent impregnation with chemical reagents, and activation [27]. When potassium hydroxide (KOH) is involved in pyrolysis, the reaction unblocks the clogged pores and widens small channels [28]. Moreover, KOH in the molten state can also erode new channels, which helps to expand the surface area of biochar. Meanwhile, KOH may also interact with oxygencontaining functional groups in biomass, which enhances the Cd adsorption capacity [29-35].

Regardless of the aforementioned methodologies, no unified standard for the optimal concentration of KOH exists. Yang et al. adopted a 5% KOH solution during the impregnation pretreatment phase of cedar sawdust biochar [36]. Xiong et al. fabricated AC by immersing peanut shell powder in KOH solutions with varying concentrations (ranging from 1 mol/L to 6 mol/L) [37]; nevertheless, their research paper failed to expound upon the influence of concentration on AC. Ghiffari et al. initially activated water hyacinth through the application of KOH, with the KOH concentrations set at 25%, 30%, and 35%, and subsequently subjected it to carbonization; the experimental results manifested that a high concentration led to a large surface area of the AC [38]. Zhu et al. synthesized AC via a one-step approach; when the mass ratio of the activator (KOH) to biomass reached 4 in the KOH aqueous solution, the micropores and adsorption capacity of the prepared AC were maximized [39]. Marques et al. exploited KOH (with a concentration of 85%) and biomass at a ratio of 1:3 to activate apple branches and then dissolved them in 5 cm³ of water [40]; however, the precise KOH concentration in the actual activation process remained indeterminate. Perdana et

al. investigated the effect of KOH solution concentration on the performance of palm shell activated carbon capacitor electrode materials [41].

With respect to the preparation methodologies, the majority of biomass AC is typically completely pulverized in the initial phase of preparation. Subsequently, deionized water is employed for cleaning purposes, followed by pH adjustment through the application of acid in the terminal stage before the adsorption experiments. Empirical findings indicate that a high concentration of KOH is conducive to the formation of biochar pore structure. However, such a condition may cause the degradation of the skeletal structure of biochar, which is unfavorable to its subsequent large-scale application. Concurrently, the pulverizing steps in the traditional carbon process increase the complexity associated with the washing process and wastewater disposal due to the increased volume, which potentially imposes an environmental liability.

Based on the preceding analysis, Cd²⁺ from the aqueous phase may be successfully removed using peanut shells as the raw material (RM) in conjunction with KOH as an activator for the production of biochar at 600 °C during pyrolysis. Nevertheless, several issues remain, and they are expounded upon as follows: (1) With KOH serving as an activator, the impregnation concentration exhibits a lack of consistency across diverse literature. (2) Considering that OH⁻ plays a facilitating role in the precipitation of Cd²⁺, the biochar prepared in the aforementioned studies typically undergoes procedures, such as pickling or washing, in the final stage to remove ash residues. Subsequently, pH is meticulously adjusted to a neutral state before conducting heavy-metal adsorption experiments. However, when contemplating the direct implementation of heavy-metal adsorption within wastewater environments (especially in acidic environments), the indispensability of the aforementioned pH adjustment step becomes a subject worthy of in-depth investigation. (3) The economic aspect also warrants attention. The expenditure associated with prepulverized biomass to a particle size of 1 mm or below 3 mm is relatively substantial [33, 38]. By contrast, the cost incurred during the post carbonization stage is relatively low. This cost reduction is mainly due to changes in material properties.

Therefore, this study aims to experimentally explore the feasibility of directly adsorbing Cd²⁺ from aqueous solutions using biochar prepared from peanut shells. Peanut shell biochar was prepared using a low-concentration KOH impregnation process, and initially measured its basic properties, such as appearance, morphology, and industrial analysis. Then, the effect of sieving on the removal rate of Cd²⁺ (Cd-RR) was evaluated, and the optimal KOH impregnation concentration and biochar feed amount were further determined. Moreover, based on reasonable inferences made from existing literature on the possible adsorption mechanisms of peanut shell AC, a discussion on the economic value of the new method was provided. This study is important in promoting the use of peanut shells to remediate Cd²⁺-contaminated water bodies, which effectively simplifies the process and reduces cost inputs.

2. Materials and methods

2.1 Materials

Samples of peanut shells (Tianfu 11 variety), which were sourced from Tuping Village, Xinmin Town, Qianwei

County, Sichuan Province, were obtained in 2018 during the Second National Agricultural Pollution Source Census in China. Following the Chinese national standard GB/T212-2008, a proximate analysis of the peanut shells was carried out, and the results are presented in Table 1.

2.2 Reagents and instruments

The main reagents included KOH (Analytical Reagent, Fangzheng Reagent Factory, Beichen District, Tianjin) and cadmium chloride (CdCl₂, Analytical Reagent, Tianda Chemical Reagent Factory, Dongli District, Tianjin), and nitrogen (N₂, 99.9%, Xinxiang Beipu Special Gas Factory). Laboratory water refers to purified water.

The main pieces of equipment included a vacuum hightemperature tube furnace (OTF-1200X-L, HF-KEJING Materials Co., Ltd.), an electric heating air-blowing drier (101-3AB, Tianjin Taisite Instrument Co., Ltd.), a scanning electron microscope (SEM) (Quanta 200, Field Electron and Ion Company), an analytical balance (BSA113S-CW, Mettler-Toledo International Inc.), a constant-temperature oscillating shaker (ZWY-2102C, Shanghai Zhicheng Analytical Instrument Manufacturing Co., Ltd.), a centrifuge (H1850R, Hunan Xiangyi Laboratory Instrument Development Co., Ltd.), an inductively coupled plasma atomic emission spectrometer (ICP, Optima 2100DV, PerkinElmer Inc.), a pH meter (pHS-3C, Shanghai Jingci Instrument Co., Ltd.), and a fully automatic specific surface area and micropore analyzer (JWGB-BK-112, Beijing JWGB Sci. & Tech. Co., Ltd.).

2.3 Preparation and physicochemical testing of biochar

Following rinsing with purified water, the RMs were dried at 105 °C and then packaged for subsequent application. Peanut shells were accurately weighed, and KOH solutions (5%, 10%, and 15% concentrations) were prepared. The shells were then immersed in the solutions at a solid-toliquid ratio of 1:5 for 4 h. During immersion, the mixture was stirred every 20 min. Afterward, the samples were left to stand for 6 h in a cool, shaded environment and then dried in an oven at 75 °C for 8 h. Finally, the dried shells were transferred to crucibles and introduced into a vacuum hightemperature tube furnace under a nitrogen atmosphere (0.1 MPa, 200 mL/min flow rate). The material was heated up to 600 °C at a heating rate of 10 °C/min. Afterward, the temperature was kept constant for 2 h. Next, natural cooling was permitted for 2 h until room temperature. As a result, the unpretreated peanut shell biochar (control group, designated as CK) and the biomass-AC treated with KOH solutions at the concentrations of 5%, 10%, and 15% (denoted as KA, KB, and KC, respectively) were obtained. Further, proximate analysis and tests of pore structural parameters were performed on the carbonaceous specimen.

2.4 Experiment on Cd removal by biochar

The CdCl₂ solution of 100 mg/L was prepared. Then, the unwashed biochar prepared by impregnation with diverse KOH impregnation concentrations was retrieved and mixed with the 50 mL CdCl₂ solution for the adsorption process. Adsorption was carried out at a stirring rate of 200 r/min and a temperature of 30 °C for 12 h. Thereafter, the mixed solution was centrifuged at 9000 r/min for 15 min to obtain the supernatant. Subsequently, an ICP spectrometer and a pH meter were used to analyze the concentration and pH, respectively.

CK and KB were selected as research targets, and they were sieved using a 140-mesh sieve. Subsequently, the

capabilities of biochar to remove Cd^{2+} before and after sieving were evaluated and tested. Thereafter, based on the optimal results obtained, a test was conducted on the Cd-RR at different feed quantities (1.0, 2.0, and 3.0 g/L) for all AC additions. The Cd-RR was calculated using Equation (1):

$$Cd-RR = \frac{(c_0 - c_t)}{c_0} \times 100\%$$
 (1)

where c_0 and c_l represent the initial and final concentrations of Cd²⁺ (mg/L) in the solution adsorbed by biomass-AC, respectively.

The overall Cd-RR after sieving was n times that before sieving:

$$n = \frac{\text{Cd-RR}_1}{\text{Cd-RR}_0} \frac{m_1}{m_0}$$
(2)

where Cd-RR₀ and Cd-RR₁ represent the removal rates of Cd²⁺ of biochar before and after sieving, respectively. m_0 and m_1 represent the weights of biochar before and after sieving, respectively (g).

According to the results of Cd-RR before and after sieving of the selected biochar, the decision on whether to adopt the sieving scheme was finally made. Additionally, the correlation tests between the feed amounts of AC (1.0, 2.0, and 3.0 g/L) and the removal rates were carried out.

3. Results analysis and discussion

In accordance with the experimental scheme, the research team first observed the surface morphology and carried out a proximate analysis of the prepared biochar products. Subsequently, the team tested the Cd-RR and analyzed the results.

3.1 Appearance morphology characterization and proximate analysis of carbon products

3.1.1 Appearance morphology characterization of biochar

Fig. 1 presents the morphological comparison of the biochar prepared with different methods. For CK, its shape remained intact, characterized by high hardness, low brittleness, negligible variation in thickness, and a relatively low carbon yield. By contrast, the biochar pretreated with KOH solution experienced substantial morphological deformation and featured low hardness and high brittleness. However, the carbon converted directly from raw peanut shell displayed the state of the original peanut shell.

Upon grinding of the three biomass-AC products, all the carbon powder had a strong tendency to stick to the inner wall of the pot. This phenomenon comprehensively indicates that the surface of the biomass-AC prepared with KOH is abundant in charges [42].

3.1.2 Proximate analysis and pore structural characteristics

(1) Proximate analysis

Table 1 shows that the ash content in the CK exceeded that in the RM. The main reason is that during biochar production, with continued decomposition and volatilization,

ash was mainly retained in biochar [43]. However, given the dehydration and etching effects of KOH, a large amount of ash in the carbon pores could be removed. With the increasing concentration of KOH, the ash content of the impregnated biochar gradually decreased. Meanwhile, in an alkaline environment, KOH reacted with the organic constituents present in peanut shells, which resulted in the generation of gaseous products and a successive diminution in the amount of volatile matter [44].



Fig. 1. Appearance observation of carbon products. (a) CK. (b) AC after 10% KOH activation

(2) Characteristics of pore structure

As depicted in Table 2, remarkable enhancements were observed in the specific surface area, micropore surface area, total pore volume, and micropore volume of biochar before and after KOH impregnation pretreatment. Concurrently, a

 Table 2. Specific surface area and pore size test of biochar

Sample type	Specific surface area (m²/g)	Microporous surface area (m²/g)	Total pore volume (cm ³ /g)	Micropore volume (cm ³ /g)	Mean pore size (nm)
CK	18.25	10.22	0.01235	0.005376	13.68
KA	402.46	348.25	0.5625	0.1234	2.95
KB	484.16	403.57	0.5954	0.1345	3.36
KC	521.73	442.25	0.6218	0.1542	3.56

3.2 Removal of Cd by biochar

3.2.1 Removal of Cd of biochar before and after sieving

To explore whether grinding and sieving can enhance the Cd-RR, CK and KB were selected for investigation. The sieved biochar was designated as Y (the screening oversize). The Cd removal performances of the biochar samples before and after sieving were assessed at a feed amount of 2.0 g/L. The results are exhibited in Table 3. A substantial improvement was observed in the removal capacity of biochar for Cd²⁺ in solutions before and after KOH activation, with the Cd-RR exceeding 98.76%. Following the grinding and sieving processes, the mass ratio of sieving reached 56.27% of the initial mass. Concurrently, the removal capacity of CK for Cd²⁺ rose from 2.03% to 9.62%. The primary cause was the limited specific surface area of raw carbon. However, following the grinding process, this area increased significantly.

 Table 3. Cd-RR of CK and KB before and after sieving

Sample type	Final Cd ²⁺ concentration (mg/L)	Cd-RR (%)	рН
CK	58.4783	2.03%	6.49
CK-Y	53.9478	9.62%	6.67
KB	0.7402	98.76%	9.52
KB-Y	0.2746	99.54%	9.72

In general, the Cd-RR after sieving increased by 2.67 times that of the initial level, although the overall removal efficiency remained unsatisfactory. After the grinding and sieving of KB, the increase in its removal capacity for Cd^{2+} compared to the untreated state was 0.78%. This finding implies that KB had good structural consistency and

considerable reduction was observed in the average pore size. With the elevation of the KOH concentration, more KOH engaged in the reaction with the peanut shell, which activated an increased number of sites. The etching intensity and reaction depth of KOH on the surface of peanut shells gradually intensified, which persistently facilitated the generation of novel micropores on their surface and the expansion of pre-existing micropores. As shown in Table 2, the micropores of AC following KOH pretreatment were relatively diminutive, which aligns with the results in relevant literature [24, 45]. Moreover, the mesopore volume constituted the principal pore structure of the KOH-AC.

 Table 1. Proximate analysis of RMs and biochar prepared under different KOH impregnation concentrations on airdried basis

Туре	Moisture (%)	Ash (%)	Volatile (%)	Fixed carbon (%)
RM	5.37	9.42	71.25	13.96
Unactivated biochar (CK)	2.15	27.48	11.25	59.12
AC activated by KA	2.35	21.54	10.66	65.45
AC activated by KB	2.48	18.08	9.47	69.97
AC activated by KC	2.57	16.28	8.02	73.13

contained less residual ash. However, the mass loss after sieving was approximately 38.12%. Calculated using Equation (2), the overall Cd-RR was 62.37% of that before sieving.

The surface morphologies of the screening oversized biochar products (CK-Y and KB-Y) and screening undersized (denoted as S) CK and KB biochar products after grinding and sieving were observed using a SEM (Fig. 2). The number of debris particles attached to the surfaces of KB-S and CK-S was greater than those of KB-Y and CK-Y. The carbon fiber framework of CK was relatively intact and featured fewer micropores. On the other hand, KB presented a rougher appearance, with a thinner shell layer of the peanut shell. In addition, KB contained more microscopic pore channels that were distributed across the entire peanut shell, and its framework underwent folding deformations. The explanations for these phenomena are as follows: First, KOH exhibited corrosive characteristics. Subjected to alkaline etching, the surface of biochar became rough, and more voids were formed. Second, during high-temperature pyrolysis, alkali reacted with functional groups, which gave rise to the release of CO, CH₄, H₂, and H₂O while generating heat. As the high-temperature gases escaped, small voids were continuously created or the existing voids were enlarged, which led to a looser internal structure of the peanut shells.

The surface of CK-Y was relatively smooth, whereas the overall surface of CK-S displayed a mountain-like bulge (Fig. 2a and 2b, respectively). A remarkable disparity existed in the maximum pore size during the comparison of the upper and lower sieves after CK was grinded and sieved, specifically 11.34 and 3.14 μ m respectively. CK-Y had a looser structure and greater pore-wall thickness than CK-S.

The chief cause was that particles with low density and loose structure were sieved out during the sieving process. Moreover, particles with an underdeveloped pore structure after carbonization, together with some ashes, such as uncarbonized minerals, were removed by sieving due to their relatively high bulk density. Hence, the adsorption of Cd^{2+} by peanut shell-derived biochar can be remarkably improved through sieving. The key factor that contributed to this improvement was the removal of large pore channels during sieving, which consequently augmented the specific surface area.

As shown in Fig. 2c and 2d, KB-S and KB-Y presented hollow characteristics on the surface, respectively, and the surface of KB-S was relatively smoother than that of KB-Y. The maximum pore sizes of the upper and lower sieves were 24.09 and 16.83 μ m, respectively, with both being characteristic of macropores. Although the particles of KB-S were finer and more fragmented, which resulted in an increase in the specific surface area to a certain extent, the growth of the specific surface area was relatively limited in comparison with the microchannels of the carbon skeleton.

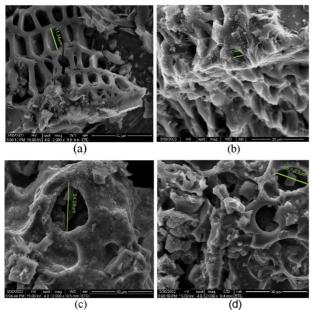


Fig. 2. SEM morphology of CK and KB on the screening oversize and screening undersize (\times 2000). (a) CK-Y. (b) CK-S. (c) KB-Y. (d) KB-S

The sieving operation was mainly aimed at the elimination of a small quantity of ash adhering to biochar. However, this process had a restricted influence on the removal capacity for Cd^{2+} and led to a reduction in the overall removal efficiency. In light of maximizing the removal efficiency of Cd^{2+} , it must be utilized directly in Cd-containing wastewater after grinding without undergoing the sieving procedure.

3.2.2 Removal of Cd²⁺ based on biochar feed amount

Adsorption experiments of Cd^{2+} by AC after grinding were directly carried out in 50 mL CdCl₂ solution (100 mg/L). When the specific surface area is not less than 1000 m²/g, the optimal amount in Cd-containing aqueous solutions is 1.0 g/L [46]. On this basis, the feed amounts in these experiments were set to 1.0, 2.0, and 3.0 g/L. The results are shown in Table 4.

(1) In this study, KA, KB, and KC exhibited the Cd-RR of more than 96% from solutions. This finding was attributed

to the fact that KOH impregnation improved the pore structure and oxygen-containing groups of biochar. The minimum Cd-RR of KA was 96.32%, whereas the Cd-RR of KB and KC exceeded 98.76%. In this experiment, the feed amount at 3.0 g/L of KB yielded the optimal removal effect, with the Cd-RR attaining 99.15%; the Cd-RR at 1.0 g/L was 98.99%.

 Table. 4. Experimental results regarding the Cd-RR using AC

Sampl e type	Feed amount (g/L)	Final Cd ²⁺ concentration (mg/L)	Cd-RR (%)	pН
	1.0	1.8683	96.87	10.06
KA	2.0	2.1966	96.32	10.43
	3.0	1.6773	97.19	10.60
	1.0	0.6029	98.99	9.12
KB	2.0	0.7402	98.76	9.52
	3.0	0.5074	99.15	9.23
	1.0	0.9252	98.45	10.29
KC	2.0	1.0565	98.23	10.73
	3.0	0.7222	98.79	11.05

(2) The Cd-RR of AC followed the order of KA < KC < KB. In comparison with KC and KB, KA possessed a less welldeveloped pore structure and the smallest specific surface area, which led to a reduction in the number of channels available for Cd²⁺ diffusion. Consequently, the number of physical adsorption sites was decreased proportionally, which resulted in a lower physical adsorption capacity for Cd²⁺. Meanwhile, the surface of KA had a smaller quantity of oxygen-containing groups capable of participating in ion exchange and surface complexation reactions with Cd²⁺. As a result, its contribution to the chemical adsorption of Cd²⁺ was also limited.

Based on the precipitation pH diagram of $Cd(OH)_2$, $Cd(OH)_2$ in KA and KC solutions were in a precipitated state, and $Cd(OH)_2$ in KB solution was in an incompletely precipitated state. The pore structure of KB was inferior to that of KC, but the Cd-RR of KB was the best, which indicates that the AC of KB had better characteristics, such as Zeta charge and functional group distribution, in chemical adsorption and need further testing.

(3) For each type of AC, the Cd-RR initially decreased and then increased as the feed amount increased, specifically in the order of 3.0 g/L > 1.0 g/L > 2.0 g/L.

When the feed amount was increased from 1.0 g/L to 2.0 g/L, the two main factors possibly contributed to the decrease in Cd-RR. First, as the feed amount rose, the interparticle distance of biochar decreased, which aggravated the agglomeration phenomenon. The adsorption sites in the agglomerated biochar became encapsulated, which caused difficulty for Cd2+ to access these sites. Consequently, the effective adsorption area was substantially diminished, which led to a decline in the adsorption efficiency. Second, the increase in feed amount resulted in a greater release of ions and functional groups from biochar into the solution, which altered the chemical environment of the solution. The increased ion concentration intensified ionic interactions and competition, which hindered the binding of Cd²⁺ to the functional groups on the biochar surface. In addition, the release of OH- from the alkaline functional groups of biochar raised the solution's pH. This increase in the solution's pH possibly led to the formation of poorly adsorbable Cd2+ hydroxy complexes, which reduced the binding affinity of Cd²⁺ to the biochar surface and ultimately lowered the Cd-RR.

The situation changed when the feed amount was increased from 2.0 g/L to 3.0 g/L. Although some sites became ineffective due to agglomeration, the substantial increase in the total amount of biochar led to a notable rise in the total number of effective adsorption sites. Meanwhile, the chemical environment of the solution stabilized. The ion concentration distribution became relatively fixed, and the mutual interference and competition among ions were remarkably mitigated. Under these conditions, the functional groups on the biochar surface could interact more efficiently with Cd^{2+} and formed a more stable adsorption structure. Therefore, the Cd^{2+} adsorption capacity was further enhanced, leading to an improvement in the Cd-RR.

(4) As the feed amount of AC increased, the pH of the KA and KC solutions showed a continuously upward trend, while that of the KB solution first increased and then decreased. The solution pH was influenced by the chemical composition of biochar itself (such as the nature and quantity of functional groups), its pore structure, and the interaction between biochar and the solution.

For KA, the KOH impregnation concentration was the lowest during the preparation process. Although a certain amount of acidic functional groups was generated during carbonization, alkaline functional groups remained predominant. As the feed amount increased, more biochar came into contact with the solution. The surface alkaline functional groups continuously released hydroxide ions, which increased the hydroxide-ion concentration in the solution. Owing to the poor pore structure of KA, alkaline functional groups concentrated on a limited surface area, and the release process remained relatively stable. Simultaneously, this poor pore structure restricted adsorption and ion exchange. The other ions generated during the adsorption process had a negligible effect on the hydroxideion concentration and couldn't counteract the pH increase resulting from the release of alkaline functional groups. Consequently, the solution pH rose continuously with the increase in the feed amount.

Since KC adopted a 15% KOH impregnation concentration, the biochar surface was endowed with a wider variety of alkaline functional groups and the fewest acidic functional groups. In addition, KC exhibited a good pore structure, which not only facilitated the full contact of basic functional groups with the solution but also promoted the diffusion of hydroxide ions in the solution and further accelerated the increase in hydroxide-ion concentration. Simultaneously, the excellent pore structure was conducive to preserving the balance of ions in the solution. The impact of acidic functional groups on the solution pH was almost negligible. Therefore, as the amount of feed continued to increase, the pH of the solution also rose persistently.

The KOH impregnation concentration used in KB was moderate, with a good pore structure. The investigated biochar not only contained abundant alkaline functional groups but also generated a relatively similar amount of acidic functional groups, e.g., hydroxyl (-OH). When the feed amount was not exceeding 2.0 g/L, the release rate of alkaline functional groups in the solution exceeded the consumption rate of acidic functional groups. At this stage, although KB had already started to adsorb Cd²⁺, its effect on OH⁻ was negligible. As a result, the pH of the solution continued to increase. When the feed amount reached 3.0 g/L, as the reaction progressed, the alkaline functional groups, after releasing a large amount of OH⁻ in the early stage, gradually decreased in number, and their capability to release OH⁻ weakened. On the other hand, during this process, acidic functional groups consumed a large amount of hydroxide ions. In addition, as Cd^{2+} was adsorbed, the concentration of Cd^{2+} in the solution decreased. Moreover, biochar possibly exhibited a stronger adsorption affinity for OH^- , which reduced the concentration of free OH^- in the solution. Furthermore, changes in the ion concentration in the solution increased the complexity of the interactions between ions, which possibly promoted the desorption of some ions originally bound to the surface of biochar and further affected the pH of the solution and ultimately led to a decrease in pH.

Therefore, the pH of the KA and KC solutions was predominantly influenced by the release of alkaline functional groups, which resulted in a continuous rise in the solution's pH. Conversely, the KB solution's pH was attributed to the combined effect of alkaline and acidic functional groups, along with alterations in biochar adsorption.

In brief, the AC prepared with 10% KOH impregnation concentration exhibited the optimal Cd^{2+} removal efficacy at the feed amount of 3.0 g/L. Subsequently, the removal efficiency at 1.0 g/L followed, with a minor difference in Cd-RR.

3.3 Discussion

3.3.1 Effect of sieving on Cd-RR

The experimental results reveal that for the activated biomass-derived AC, following grinding and sieving, the surface of AC became smooth, and the adhered ash could be effectively eliminated. Nevertheless, the removal capacity of Cd^{2+} by the sieved AC was only marginally enhanced, and the mass loss was substantial, which led to a remarkably lower overall removal efficiency compared with that prior to sieving. Hence, the grinded AC was directly employed to eliminate Cd^{2+} from water to improve the removal efficiency of heavy metals.

This study focused on the experiments using a 140-mesh sieve. In other experiments, 100- and 200-mesh [24, 47] sieves were utilized. The outcomes regarding overall removal efficiency might vary and thus warrant further investigation.

3.3.2 Optimal KOH impregnation concentration and feed amount

With respect to the procedures of KOH impregnation pretreatment of peanut shells followed by AC preparation at 600 °C, under impregnation pretreatment concentrations of 5%, 10%, and 15%, the Cd-RR of all biochar surpassed 96%. The Cd-RR was optimal when the KOH concentration was 10% and the feed amount of 3.0 g/L, followed by that observed at a feed amount of 1.0 g/L. However, from the perspective of economic performance evaluation, in industrial processing, 5% KOH impregnation pretreatment with a feed amount of 1.0 g/L might be the most cost-effective option, which requires specific analysis according to particular project circumstances.

3.3.3 pH of the solution and Cd-RR

Conventionally, the fabricated biochar was subjected to pickling or multiple washings, and the pH was adjusted to neutral. The objective was to eliminate the unreacted KOH and ash within biochar to guarantee the integrity of pore channels. Based on the outcomes of this experiment, unwashed KA, KB, and KC could remove more than 96% of Cd^{2+} from the solutions.

Nevertheless, only the biochar prepared with a 10% KOH impregnation concentration exhibited a relatively low pH when the treated solution was directly used (Tables 3 and 4). According to the optimal Cd²⁺ removal process, the pH after solution treatment was 9.23, and subsequent water treatment operations were also requisite.

3.3.4 Analysis of the mechanism underlying Cd removal

Based on the current research findings, the potential adsorption mechanism was explored. In this experiment, multiple factors synergistically influenced the adsorption process. As pointed out in the literature [47,48], the removal of Cd^{2+} by AC involved various mechanisms, including electrostatic attraction, ion exchange, physical adsorption, complexation, and precipitation.

The ionic diameter of Cd^{2+} in an aqueous solution was approximately 0.19 nm. The average pore diameter of the unactivated peanut shell biochar exceeded 2 nm [43], and that of peanut shell AC used in this study was larger. Its favorable pore structure and high specific surface area provided diffusion channels, which enabled Cd^{2+} to penetrate the interior of biochar [49]. This favorable pore structure and large specific surface area not only increased the number of physical adsorption sites but also offered a location for ion exchange, complexation, and other reactions [50].

The oxygen-containing groups on the peanut shell biochar before and after modification were crucial for chemical adsorption [51]. These groups could undergo ion exchange and surface complexation with Cd²⁺. During ion exchange, ions on the biochar surface were exchanged with Cd²⁺ for fixation. Surface complexation enhanced the binding force by forming chemical bonds. The alkaline nature of biochar endowed its surface with a large amount of charge, which generated electrostatic attraction with Cd2+. This electrostatic attraction caused Cd²⁺ accumulation on the biochar surface, which created favorable conditions for subsequent adsorption. When the solution pH was high, Cd2+ reacted with OH- to form a precipitate, which may adhere to the biochar surface and increase the adsorption capacity. Ionic exchange occurred between the ash in biochar and Cd²⁺, and a complexation reaction transpired between the hydroxyl functional groups and Cd2+, which jointly promoted the adsorption process.

According to literature [24], the adsorption process of modified biochar is mainly governed by chemical adsorption, which may also be applicable to Cd^{2+} adsorption by peanutshell AC. The chemical reactions involving oxygen-containing groups, ash components, and hydroxyl functional groups in peanut shell biochar may play a pivotal role in

Cd²⁺ removal. Compared with physical and electrostatic adsorption, these chemical reactions can immobilize Cd²⁺ more stably, which improves adsorption efficiency and stability. Nevertheless, further research is required to clarify the dominant role of chemical adsorption and the contributions of various chemical reactions.

This study focused on the final practical effect of the direct biochar application after KOH impregnation. Nevertheless, owing to the constraints of the research focus, no relevant investigations were conducted regarding the Zeta potential, adsorption isotherm, and kinetic model experiments. As a result, the detailed adsorption mechanism could not be fully analyzed, and thus, related research will be carried out in the future.

3.3.5 Economic analysis of the direct application method

Compared with the traditional method, the direct application method has advantages in terms of the RM cost, preparation process, and product benefit for the removal of Cd^{2+} in aqueous solutions (Table 5).

(1) KOH cost Although peanut shells and KOH were both required, KOH could also take part in certain reactions during the subsequent removal of Cd^{2+} . Theoretically, this scenario can enhance the utilization efficiency of KOH to some extent and relatively decrease its overall cost under the same output effect.

(2) Preparation process cost. The energy consumption cost in the activation stage is comparable to that of the traditional method. Nevertheless, in subsequent Cd^{2+} removal and solution pH adjustment, the application of chemical reagents becomes more targeted. An appropriate amount of alkali solution may be added to delicately adjust the final pH. Compared with the traditional approach involving pickling and wastewater treatment, the method used in this experiment can economize on the cost of acid reagents and cut down the cost of wastewater treatment.

(3) Profit analysis The biomass-AC processed by traditional methods is applicable to a broader range of fields, including soil improvement, environmental remediation, and other conventional scenarios with stringent pH requirements. It enjoys a wide market coverage and can generate relatively stable sales revenue. However, the new method holds a greater allure in projects that require Cd-contaminated water treatment. It may reap higher value-added benefits, particularly in the environmental protection treatment market which has a substantial demand for Cd-containing wastewater treatment along with a specific fee standard, which renders its benefits important.

Overall, in regard to the treatment of Cd²⁺ waste liquid, the new method features a relatively low cost and great application advantages.

Table 5. Economic comparison between the traditional and direct application methods

Comparison projects	Traditional method (KOH activation-pH adjustment by acid washing before Cd ²⁺ removal)	Direct application method (KOH activation-pH adjustment after Cd ²⁺ removal)
KOH cost	Relatively high (KOH is solely utilized for activation, and the quantity is within the conventional range)	Relatively low (the utilization efficiency of KOH is enhanced, or the amount is marginally decreased under the same output level)
Preparation process cost	High (involving pickling and wastewater treatment procedures, with a large number of equipment and reagents, considerable energy consumption, and high treatment costs)	Relatively low (without the necessity for complex pickling and wastewater treatment processes, accompanied with slightly low costs in terms of reagents, equipment, and energy consumption)
Profit analysis	It is relatively stable and encompasses wide application fields, such as soil improvement and conventional environmental restoration.	The added value is potentially higher and more targeted (in the field of cadmium-containing wastewater pollution control).

4. Conclusions

In this study, from the perspective of industrial application, peanut shells from agricultural waste were used as RM to remove Cd^{2+} in solutions. The approach of KOH

impregnation activation followed by pyrolysis carbonization was utilized to directly carry out the removal experiment of Cd²⁺ residual rinse. Finally, the following conclusions were obtained:

(1) The highest overall Cd-RR was achieved when the biomass-AC was directly utilized without pH adjustment or washing treatment. The biochar prepared at a 10% KOH impregnation concentration exhibited the best Cd^{2+} removal capability, which reached 99.15% when the feed amount was 2.0 g/L under direct use. Next, Cd-RR was 98.99% when the feed amount was 1.0 g/L. One of these two feed quantities can be selected in accordance with specific requirements and performance expectations. However, from an economic perspective, a feed amount of 1.0 g/L was recommended.

(2) Although sieving can remove ash from AC and slightly enhance its performance, it also results in tremendous mass loss, which ultimately reduces the overall Cd^{2+} removal capacity. Under standard conditions, the grinded AC can be directly employed for the pollution remediation in Cd^{2+} -containing wastewater. Thus, the utilization of sieved AC is advisable only when performance is the primary consideration. Conversely, for common biochar materials, sieving represents a prompt approach to enhancing their performance under urgent scenarios.

(3) In the context of Cd-containing wastewater treatment, this method demonstrated an enhanced adaptability. With minimal reduction in the Cd-RR, when

compared in terms of RM cost, preparation process, and benefit analysis, it exhibits advantages over the traditional biochar preparation using KOH for water restoration. Nevertheless, given the elevated pH of the treated aqueous solution, a preproject evaluation is requisite to preclude other environmental risks arising from the strong alkalinity of the solution. This method is recommended for application in flowing water bodies.

The study can effectively simplify the process of treating Cd-containing wastewater using peanut shells. In future studies, this method can be applied to the treatment of acidic solutions containing heavy metals by using other biomass, and the specific mechanism underlying chemical adsorption needs further research. Moreover, the research conclusions provide novel directions for enhancing the utilization of waste biomass in the remediation of heavy metal water pollution.

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