

Review

Adsorption of Lead from Aqueous Solution by Biochar: A Review

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Abstract: Industrialization has resulted in the discharge of a certain amount of lead (Pb) from industrial sources causing damage risk to water quality and human health. Adsorption is an effective technique to remove Pb, and biochar has been widely studied owing to its advantages of low cost and high adsorption capacity. This review summarizes the influence of raw materials and modification methods on the adsorption capability of biochar. The adsorption isotherms and kinetics of biochar were summarized, and the main Pb removal mechanisms were studied systematically. In addition, the challenges and future perspectives were discussed comprehensively. It is expected that the review could provide insightful fundamentals for the experimental research and practical applications of biochar.

Keywords: heavy metal; lead; biochar; wastewater; adsorption



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1. Introduction

Since the last century, the intensification of industrialization has put great pressure on the environment [1]. Automobile maintenance, tetraethyl manufacturing [2], refining process, and battery manufacturing process have produced a large amount of toxic lead (Pb), which will pollute the water [3]. Pb is a toxic and harmful element with high molecular weight, and it has the most abundant global distribution among heavy metals [2,4,5]. Besides, Pb is capable of accumulating in the human body, leading to serious health problems including cancer, anemia, renal insufficiency, permanent brain damage [6], and extreme mutations [7]. Recently, Pb pollution has become a major problem of water pollution in developing countries [8]. Therefore, removing Pb from wastewater is an important and urgent task to ensure human life and health [9].

In recent decades, a variety of techniques have been developed to remove Pb from wastewater, such as ion-exchange [10], membrane processes [11], chemical precipitation [12], filtration [13], electrocoagulation [14], coagulation [15], and adsorption [16–19]. Among these methods, adsorption is one of the most common removal technologies and has attracted wide attention owing to its advantages of low cost, simple process, and high removal efficiency [20–22]. Many adsorbents have been used to remove heavy metals from aqueous solutions. Traditional adsorbents may be unpopular due to their high production and regeneration costs [23], while lower-cost biochar is an effective material for removing Pb from water and has received extensive attention [24]. Adsorption features have been very recently exploited not only for environmental remediation by simple adsorption but also for sensing and catalytic applications [25–27].

Biochar, a porous carbonaceous material, is produced by the pyrolysis of biomass in an oxygen-limited environment [28]. Considering that biochar originates from the ‘Terra Preta’ soils in the Amazon region, the application of biochar for soil improvement was extensively investigated. Recently, biochars have been widely used in practical environments as air filters, fuels, building materials, and adsorbents [29]. Notably, biochar is proved to be a promising adsorbent for Pb removal in wastewater [30]. Common materials for biochar production include crop residues [31–33], wood, animal manure [34], and sewage sludge [35]. The adsorption capacity of biochar with different raw materials (sludge, cow dung, corn stalk, and willow) for heavy metals was explored, and it was found that corn stalk biochar had the best adsorption effect, which might be attributed to its larger specific surface area and excellent pore structure [36]. Biochar has a porous structure, large specific surface area, and abundant surface functional groups, which enables it to efficiently adsorb Pb from aqueous solutions [37]. However, the adsorption capacity of pristine biochar may be lower than that of conventional adsorbents [38]. For instance, the maximum Pb adsorption capacity of pristine straw biochar produced at 500 °C was 165 mg·g^{−1} [39]. Therefore, chemical or physical modifications are usually carried out to enhance the adsorption capacity of biochar [40,41]. For example, the N-doped MgO-modified corn cob biochar had an adsorption capacity of 1429 mg·g^{−1} for Pb in aqueous solution [42]. In addition, the micropore area and external specific surface area of ball-milled bone biochar were significantly increased, and the Pb adsorption capacity of ball-milled biochar was up to 558.8 mg·g^{−1} [43]. Hence, the modified biochars have considerable adsorption capacity of Pb, which can promote the application potential of biochar in wastewater treatment. At present, most of the relevant reviews are related to the summary of multiple heavy metals removed by biochar in wastewater, and there are relatively few summaries of single heavy metal. Considering the continual emergence of scientific advancement, it is necessary to systematically summarize the recent development of the Pb adsorption in wastewater by the pristine and modified biochars.

This paper focuses on the adsorption of Pb in wastewater by biochars. Important aspects such as raw materials, modification methods, and removal mechanisms are summarized. The overall purpose of this review is to provide a comprehensive and systematic analysis of the latest research results on biochar for Pb removal. The specific tasks of this work are as follows: (1) Collect relevant data about the Pb adsorption by biochar; (2) Clarify the influence of raw materials and modification methods on the Pb adsorption of biochar; (3) Explore the kinetics and isotherm models of Pb adsorption by biochar; (4) Summarize the adsorption capacity and removal mechanism of biochar; (5) Point out the shortcomings of existing research and provide guidance for Pb adsorption by biochar.

2. Feedstocks for Biochar Production

A large amount of solid waste, such as crop straw and sludge, has caused a series of environmental problems. The preparation of biochar can achieve the sustainable recycling of resources. According to the existing raw material data of biochar used for Pb adsorption, the raw materials were mainly divided into the following six categories: wood materials, agricultural wastes, animal residue, sludge, fruit peel, and other wastes. The proportions of the six raw materials are approximately 23.01%, 46.02%, 7.08%, 8.05%, 5.31%, and 9.73% (Figure 1).

Biochars prepared from different raw materials have different adsorption capabilities. The adsorption capacity of different raw material-derived biochars for Pb is presented in Figure 2. As for wood materials, the KMnO₄-modified hickory wood biochar obtained a maximum Pb adsorption capacity of 153.1 mg·g^{−1} [44]. In addition, pine sawdust biochar had an adsorption capacity up to 606.00 mg·g^{−1} [45]. Therefore, wood could be made into biochar as an effective material for heavy metal adsorption.

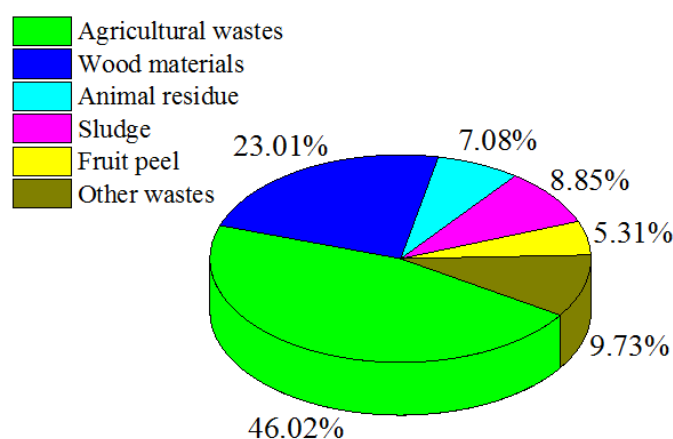


Figure 1. The proportion of raw materials for biochar to adsorb Pb in water.

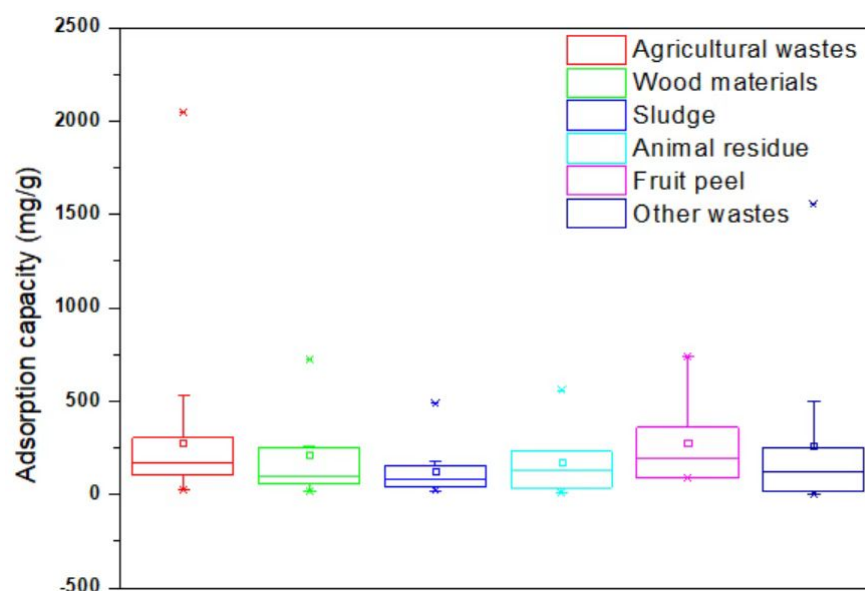


Figure 2. The Pb adsorption capacity of biochars derived from different raw materials.

As for agricultural wastes, *Medulla tetrapanacis* biochar had a high efficiency for the sorption of Pb ($1031.23 \text{ mg}\cdot\text{g}^{-1}$) [46]. Similarly, corncob-to-xylose residue biochar could efficiently adsorb Pb in aqueous solution with a maximum adsorption capacity of $1429.00 \text{ mg}\cdot\text{g}^{-1}$ [42]. There was one study that showed that the excellent adsorption capacity of tobacco stem biochar was $2047.00 \text{ mg}\cdot\text{g}^{-1}$ according to the Langmuir model [47]. Agricultural waste biochar has attracted more attention than other raw materials (Figure 1). It is a key material for biochar research and industrial application.

As for animal residue, the biochar prepared from cow bone and optimized by ball milling technology had an adsorption capacity of $558.88 \text{ mg}\cdot\text{g}^{-1}$ [43]. At the same time, biochar derived from cow manure showed a maximum adsorption capacity of about $230.00 \text{ mg}\cdot\text{g}^{-1}$. The animal residue biochar has a certain adsorption capacity, but the relative research is not enough.

As for sludge, coagulation sludge biochar had a maximum adsorption capacity of $450.58 \text{ mg}\cdot\text{g}^{-1}$ [48]. As for fruit peel, there were six related papers, and the Pb adsorption capacities were, respectively, $86.96 \text{ mg}\cdot\text{g}^{-1}$ [49], $247.10 \text{ mg}\cdot\text{g}^{-1}$ [50], $134.00 \text{ mg}\cdot\text{g}^{-1}$ [51], $88.70 \text{ mg}\cdot\text{g}^{-1}$ [52], $742.00 \text{ mg}\cdot\text{g}^{-1}$ [53], and $359.00 \text{ mg}\cdot\text{g}^{-1}$ [54]. It could be seen that the fruit peel was a suitable raw material for biochar production. The adsorption capacities of different biochars are shown in Figure 2. In addition, the different adsorption capacities are ascribed not only to the nature of the feedstock but also to the pyrolysis conditions and modification techniques.

3. Modification Methodology

Many experimental results showed that the unmodified biochar had a weak adsorption capacity for heavy metals [55]. Thus, the biochar could be modified to further improve the adsorption capacity. The surface functional groups, pore structure, specific surface area, cation exchange capacity, and other physicochemical properties could be greatly improved after modification [31].

As shown in Figure 3, there are three main methods of biochar modification: biological modification, physical modification, and chemical modification. There are many studies on chemical modification, and it has a relatively good modification effect. Therefore, the chemical modification method will be analyzed and summarized, and the physical and biological modification methods will be introduced briefly.

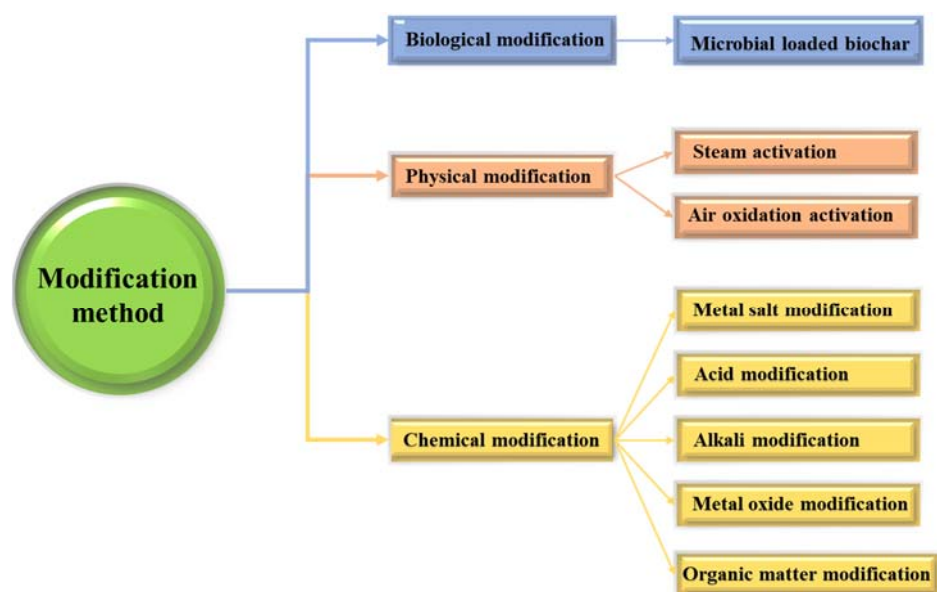


Figure 3. Biochar modification methods for adsorbing Pb.

3.1. Biological Modification

The well-developed pore structure and acid-base buffering capacity of biochar are beneficial to the growth of microorganisms. Biologically modified biochar is mainly used to load suitable microorganisms, which has achieved synergistic effects in both biochar adsorption and microbial adsorption, but suitable microorganisms need to be screened. Besides, anchoring proteins on the cell surface have metal-responsive motifs that can be specialized for metal adsorption and dissociation [56]. Wang et al. studied the Pb adsorption by biochar-loaded UV-mutant *Bacillus subtilis*, and a higher Pb adsorption capacity was achieved for the modified biochar [57]. The adsorption amount of Pb on the original biochar was about 150 mg/g, and the adsorption amount increased to 166.8 mg/g after loading bacteria, which might be attributed to the synergistic effect of biochar and bacteria. In addition, the microbial surface contains a large number of adsorption sites, which improves the adsorption effect of biochar on Pb [58]. However, considering that bacteria have strict requirements on reaction conditions, the Pb removal efficiency of bacterium-modified biochar would be affected during the treatment of industrial wastewater.

3.2. Physical Modification

Physical modification methods mainly aim to optimize the physical properties of biochar (pore size, specific surface area, etc.). Common physical modification methods include steam activation, air activation, and ball-milling. Steam activation is a relatively common physical modification method, which will increase the specific surface area and pore size of biochar after activation. For example, the steam-activated biochar had a higher adsorption capacity for Pb due to the large surface area [59]. Air oxidation also has a

good modification effect. For instance, the Pb adsorption capacity increased from 2.50 to 44.00 mg·g⁻¹ after the air-oxidized operation [60]. Furthermore, the biochar modified by ball milling has excellent heavy metal adsorption capacity, with the advantages of environmental friendliness and wide adaptability [61]. The bone-derived biochar showed a larger specific surface area and a better pore structure after ball-milling, which promoted the adsorption of Pb [43]. However, the separation of ball-milled biochar after Pb adsorption should be paid more attention during the practical application.

3.3. Chemical Modification

Chemical modification is mainly divided into the following categories: metal salt modification, acid modification, alkali modification, metal oxide modification, and organic matter modification.

3.3.1. Metal Salt Modification

The most used modifier is metal salt, which has an excellent modification effect on the pore size, the specific surface area, and the metal ions on the biochar surface. In addition, magnetic substances, such as iron, can enhance the magnetic properties of biochar, which helps to separate the biochar after adsorption.

The co-pyrolysis of magnesium-containing compounds, such as MgCl₂, with biomass can increase the specific surface area, active sites, and surface charge of biochar [62]. For example, the adsorption capacity of MgCl₂-modified biochar was 20 times higher than that of unmodified biochar [40]. The content of carboxyl, lactone, and phenol functional groups of modified biochar increased by 0.50, 0.86, and 0.06 mmol/g compared with the original biochar. The magnetization of biochar was a popular modification method. For example, magnetic cane biochar contained 18.40% oxygen, which could swell in water and allowed Pb to be adsorbed on the biochar [63]. Potassium permanganate (KMnO₄) was a kind of metal salt with good modification effect owing to its strong oxidation property. KMnO₄ modification promotes the formation of hierarchical structure, while increasing the specific surface area of biochar and optimizing the pore structure [64]. The Pb adsorption capacity of hickory wood biochar modified by KMnO₄ was 153.10 mg·g⁻¹, which was significantly higher than the original biochar, because the modified biochar had more surface oxygen-containing functional groups and larger surface area than the original biochar [44].

3.3.2. Acid Modification

Acid-modification is mainly used to increase the content of oxygen-containing functional groups to increase the adsorption ability of the biochar. Currently, phosphoric acid (H₃PO₄) and sulfuric acid (H₂SO₄) are the main acids for biochar modification.

H₃PO₄ could activate biochar to form –P=O and –P=OOH functional groups [65]. For example, orange peel biochar modified by H₃PO₄ had successfully introduced oxygen and phosphorus functional groups [52]. In addition, H₃PO₄ modification was beneficial to improve the phenolic group content and alkalinity of biochar, and the Pb adsorption capacity of the modified biochar was 5.41 times higher than unmodified biochar [66]. The carboxyl content on the surface of modified biochar increased significantly, which improved the adsorption capacity of biochar for heavy metals [67].

H₂SO₄ is corrosive and oxidizing, which contributes to the formation of pore- and oxygen-containing functional groups on biochar. H₂SO₄ will sulfonate with the aromatic hydrocarbons and introduce –SO₃H on the biochar [68]. Li et al. found that the lignin biochar modified by H₂SO₄ could introduce highly acidic oxygen-containing groups, and the maximum Pb adsorption capacity reached 679.00 mg·g⁻¹ [69]. Meanwhile, the biochar modified by H₂SO₄ had strong complex ability and high electronegativity, and a large amount of –OH and –COOH were generated on the surface of biochar during the sulfonation process. In addition, the Pb adsorption capacity for biochar was up to 191.07 mg·g⁻¹ within 5 min [70].

3.3.3. Alkali Modification

Alkali modification facilitates the optimization of the pore structure and specific surface area of biochar. The biochar surface has more adsorption sites by improving the pore structure and increasing the specific surface area.

Common alkaline modification reagents for biochar are potassium hydroxide (KOH) and sodium hydroxide (NaOH). KOH modification method could increase the content of functional groups containing O and N on the surface of biochar, and promote ion exchange and surface complexation reaction [71]. For example, Herath et al. found that modified biochar had a higher adsorption capacity than unmodified biochar, because the specific surface area of KOH-activated biochar was doubled compared with the raw biochar [72]. Compared with the pristine biochar, NaOH-modified biochar has a better mesoporous structure and higher adsorption capacity [73]. In addition, Ding et al. demonstrated that the Pb adsorption capacity of NaOH modified biochar was 2.6–5.8 times higher than that of the original biochar, because cation exchange capacity and the surface area of the modified biochar were significantly improved [74].

3.3.4. Metal Oxide Modification

Metal oxides loaded on biochar can change the physicochemical properties of biochar, such as the elemental composition. The Pb adsorption capacity of biochar modified by metal oxide could be improved [75]. MgO-modified biochar or MgO-biochar composites have certain application prospects in the field of heavy metal adsorption [53]. At the same time, Zhang et al. found that the maximum Pb adsorption capacity of the MgO-coated biochar was 558.00 mg·g⁻¹ when the maximum Mg content in the biochar was 10.10% [53]. On the one hand, MgO modification increases the surface area of biochar, and the higher surface area facilitates the adsorption/removal of Pb [76]. Additionally, the presence of the MgO coating significantly enhances the buffering capacity of the solution [77].

3.3.5. Organic Matter Modification

Organic matters were used to optimize the functional groups on biochar. The impacts of organic matter modification on the physicochemical properties of biochar are mainly reflected in two aspects. Firstly, the organic matter had an influence on the content of the functional groups. Chitosan is a surface modifier that can modify biochar to introduce a large number of oxygen-containing functional groups [78]. For example, Deng et al. proved that chitosan-pyromellitic dianhydride-modified biochar had more surface functional groups than the original biochar, so it had better Pb adsorption capacity [79]. The adsorption capacity of modified biochar for Pb improved by 10% compared with the original biochar, and the organic matter-modified biochar had a certain selectivity for heavy metal adsorption in wastewater. Furthermore, the biochar modified by thiourea could introduce C-O, C=O, and C=S, which could adsorb Pb through coordination and ion exchange, thereby thiourea-modified biochar increased the adsorption capacity of Pb(II) by 32% compared with the pristine biochar [80]. The second aspect is that organic matter influenced the elemental composition of biochar. The ammonium polyphosphate-modified biochar had more abundant N and P functional groups than unmodified biochar, so the Pb removal performance of modified biochar (723.60 mg·g⁻¹) was improved compared to unmodified biochar (264.20 mg·g⁻¹) [81].

3.4. Comparison of Different Modification Methods

Among these modification methods, metal oxide and metal salt modification achieved the most significant improvement in the Pb adsorption capacity of biochar, and the rise ranged from 42% to 600% [81–83]. The Pb adsorption capacity of Mg-modified coconut shell biochar was 20 times higher than that of the pristine biochar [40], and the N-doped coupling MgO-modified biochar derived from corn-cob-to-xylose residue exhibited a remarkable Pb adsorption capacity of 1429 mg/g [42]. Besides the enhancement in the surface morphology of biochar, the addition of metal oxide and metal salt could promote the ion exchange

capacity of biochar, thus boosting the Pb adsorption capacity. However, the organic modification method has limited effect on improving the Pb adsorption capacity of biochar. The adsorption capacity of chitosan-modified biochar for Pb was only increased by 10% compared with the pristine biochar. Notably, the physical modification (e.g., air activation and ball-milling) has a good potential to optimize the Pb adsorption capacity of biochar by modifying the physicochemical properties. The Pb adsorption capacity of the shredded wood-derived biochar modified by mild air activation was 17 times higher than that of the raw biochar, since the modified biochar had a higher specific surface area and more abundant oxygen-containing functional groups on the surface [61]. The significant improvement in the micropore area and external specific surface area of bone biochar were also observed after ball-milling, and the Pb adsorption capacity of the ball-milled biochar increased by 64.6% compared with the pristine biochar [43]. In contrast, the improvement effect of bio-modified biochar on Pb adsorption is limited, even though the size distribution of pores in biochar provides suitable habitats for many microorganisms.

Hence, the specific chemical modification techniques such as metal oxide and metal salt modification show the greatest potential in terms of the improvement in Pb sorption capacity, while the negative effects of chemical modification including the high cost and the secondary pollution should be concerned during the practical application. Considering the simple operation procedure, good economic feasibility, and environmental friendliness of the physical modification, it could be an alternative technique to promote the Pb sorption capacity of biochar, and the enhancement efficiency could be further improved. In comparison with the aforementioned modification techniques, the biological modification showed a relatively poor potential in the practical application owing to the limited Pb adsorption capacity and strict application conditions.

4. Adsorption Isotherm and Kinetics

4.1. Adsorption Isotherm

The exploration of adsorption isotherm is very important for understanding the interaction between biochar and heavy metals. Freundlich ($q_e = K_F C_e^{\frac{1}{n}}$), Langmuir ($q_e = \frac{q_{max} C_e}{K_L^{-1} + C_e}$), and Temkin ($q_e = \frac{RT}{b} \times \ln(A_T \times C_e)$) models are often used to analyze experimental data and describe the adsorption equilibrium of Pb on biochar. According to the existing data of Pb adsorption by biochar in Figure 4, approximately 71.29% of the papers are in line with the Langmuir model [54,57,79,84–90], implying that the Pb adsorption on the surface of biochar is more like the single layer adsorption. For example, Wongrod et al. found that the adsorption isotherm of Pb by digestate biochar could be well described by the Langmuir isotherm model [91].

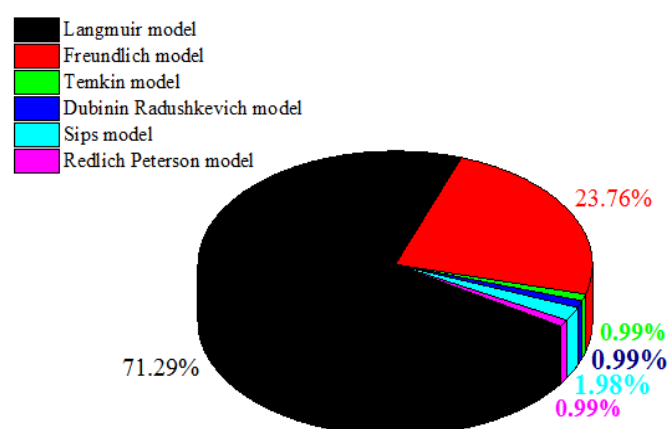


Figure 4. The proportion of different isotherm models of Pb adsorption by biochar in water.

About 23.76% of the papers are in line with the Freundlich model [59,66,69,92–97]. The heterogeneity of the biochar surface made the adsorption process more complicated,

and multi-layer adsorption might occur at sites with uneven energy distribution on the biochar [98]. Gayathri et al. used various equilibrium models to analyze the adsorption equilibrium data, and the results showed that the Freundlich isotherm was more suitable for explaining the adsorption characteristics [93]. Similarly, the adsorption of Pb in aqueous solution by nitrogen-doped magnesium oxide-modified biochar was best fitted to the Freundlich model [42]. A very small proportion of reported data conformed to other related adsorption models, such as Dubinin-Radushkevich model [75,99], Sips model [38,100], Redlich Peterson model [44], and Tempkin model.

4.2. Adsorption Kinetics

Kinetics model was used to study the adsorption rate. Three most popular kinetic models, pseudo-first-order kinetic model ($\frac{dq_t}{dt} = k_1(q_e - q_t)$), pseudo-second-order kinetic model ($\frac{dq_t}{dt} = k_2(q_e - q_t)^2$) and Elovich model ($q_t = \frac{1}{B} \ln(ab) + \frac{1}{B} \ln t$) have been used to study the adsorption of Pb on biochar [33,101].

According to the existing data of Pb adsorption by biochar in Figure 5, approximately 87.5% of the paper conforms to the pseudo-second-order kinetics model [48,54,57,59,66,69,79,84,86,88,90,100,102–111]. For example, Chen et al. applied the pseudo-first-order model and pseudo-second-order model to study the adsorption kinetics of Pb, and the results showed that the pseudo-second-order model had the best fit, revealing related adsorption processes mainly due to chemical adsorption [112]. Only 6.25% of the paper showed that the adsorption of Pb in water by biochar is in line with the pseudo-first-order kinetic model. Xue et al. found that the Pb adsorption by biochar had the best fit on the pseudo-first-order model [113]. It was assumed that the inorganic components in the biochar had low activity, and the chemical adsorption of Pb on the biochar was not obvious.

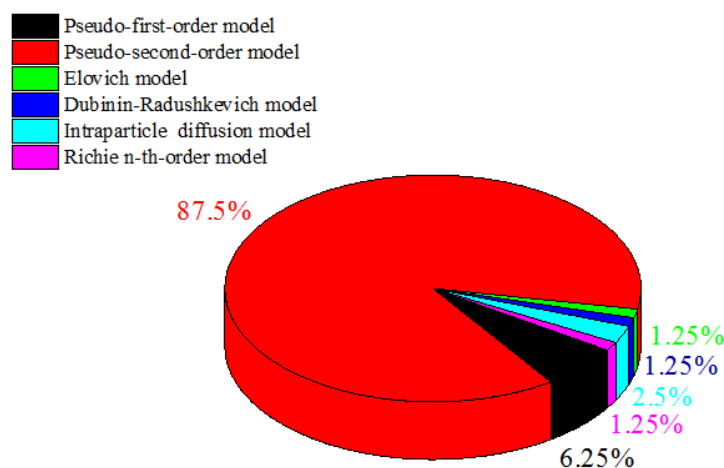


Figure 5. Proportion of different kinetic models of Pb adsorption by biochar in water.

In addition, there are two papers showing that the adsorption process of biochar conformed to both the pseudo-first-order kinetics and pseudo-second-order kinetic models. On one hand, Wang et al. discovered that both the first-order and second-order kinetic equations fitted well in describing the adsorption kinetics of Pb, which indicated that the Pb adsorption on biochar might be controlled by multiple mechanisms [114]. On the other hand, the adsorption kinetic was more in line with the pseudo-first-order model when the Pb concentration was low, and it was more in line with the pseudo-second-order model when the Pb concentration was high [115]. Thus, the Pb adsorption might be attributed to the combined effect of physical and chemical adsorption processes [116].

5. Pb Removal Mechanisms by Biochar

The Pb removal mechanisms by biochar are analyzed according to Table 1. The removal mechanisms mainly include precipitation, complexation, ion exchange, electrostatic attraction, chemical bond adsorption, and physical adsorption. The corresponding mechanisms are summarized in Figure 6.

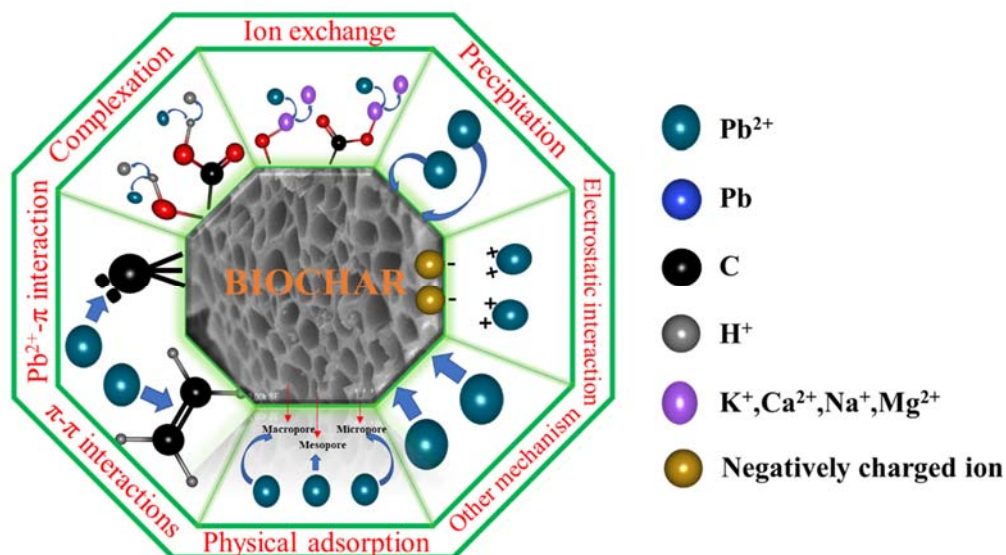


Figure 6. Mechanisms of Pb sorption by biochar.

Table 1. Information about the adsorption of heavy metal Pb in water by biochar.

Feedstock	Modifying Agent	Temperature (°C)/Residence Time (h)	Concentration Range (mg/L)	Contact Time (h)	Isotherm Model	Kinetic Model	Adsorption Capacity (mg/g)	Adsorption Mechanism	Ref.
Douglas fir	KOH	900–1000, 10 s	25–1000	1	Langmuir	Second	140.00	—	[72]
Sludge	Persulfate-zvi	600/1.5	0–100	8	Langmuir	Second	180.00	Electrostatic attraction, electrostatic outer sphere complexation, ion exchange, reduction	[112]
Raw sewage sludge	Potassium hydroxide	350/15 min	10–1000	80 min	—	Second	106.00	Precipitation	[91]
Quercus robur	FeCl ₃ and FeCl ₂	250/4	25–150	4	Langmuir	Second	63.60	Electrostatic interactions	[117]
Pine wood	MnCl ₂ ·4H ₂ O	600/1	1–300	24	Langmuir and Freundlich	First and second	47.05	Precipitation	[114]
Rice straw	—	550/2	0–1 mmol·L ^{−1}	72	Langmuir	—	0.85 176.12	Chemical complexation	[118]
Cow bone	—	600/2	0–120	24	Langmuir	Second	558.88	Surface complexation, cation exchange, chemical precipitation, electrostatic interaction, and cation- π bonding	[43]
Rice husk	β -cyclodextrin	300/2	10–300	2	Langmuir	Second	240.13	Electrostatic attraction and complexation	[119]
Palm oil sludge	—	500/0.5	0–150	4	Freundlich	First, second-	21.76	Boundary layer diffusion	[115]
Sewage sludge	—	500/0.5	0–320	6	Langmuir	First	41.20	—	[113]
Fallen leaf	Fe ²⁺ /Fe ³⁺	450/1	0–1000	2	Langmuir	Second	146.84	—	[120]
Water hyacinth	—	433/2.65	0–1000	6	Freundlich	Second	251.39	Precipitation, electrostatic adsorption, surface physical adsorption, ion exchange, and complexation of functional groups.	[121]
Cinnamomum camphor	Ultrasound-assisted alkali	450/1	50–1000	6	Langmuir	Second	98.33	Electrostatic attraction and surface complexation	[122]
Bean-worm skin waste	—	500/4	20–300	200	Langmuir	Second	62.00	Chemisorption and precipitation	[123]
Corn stalks	Nanoscale zero-valent iron, KOH	800/2	10–200	8	Langmuir	Second	480.90	Reduction reaction, complexation, and co-precipitation	[124]
Rice husk	Manganese oxide	800/3	15–250	1.5	Langmuir	Second	86.50	Electron density reduction in π -bond aromatic moieties due to the addition of -COOH	[125]
Corn cob-to-xylose residue	Nitrogen doped magnesium oxide	400/2	0–500	24	Freundlich	Second	1429.00	Ion exchange, precipitation and complexation	[42]
Date seed	HCl	550/3	—	—	Sips model	—	188.55	—	[38]

Table 1. Cont.

Feedstock	Modifying Agent	Temperature (°C)/Residence Time (h)	Concentration Range (mg/L)	Contact Time (h)	Isotherm Model	Kinetic Model	Adsorption Capacity (mg/g)	Adsorption Mechanism	Ref.
Long-root Eichhornia crassipes	—	350/20 min	1.0 mmol·L ^{−1}	50 min	Langmuir	Second	118.10	Complexation	[126]
Eucalypts leaf residue	ZnCl ₂ , FeCl ₃ and FeSO ₄	700/2	0–100	48	—	—	52.40	—	[127]
Sludge	—	550/2	200–1000	4	Freundlich	Second	30.88	Co-precipitation, complexation	[128]
Microcrystalline cellulose	Iron nanoparticles	1000	0–1500	24	Tempkin	Second	17.30	Reduction	[129]
Banana peels	—	600/2	0–600	48	Freundlich	Second	247.10	Electrostatic attraction	[50]
Rice husk	—	700/1	0–600	48	Freundlich	Second	26.70	Precipitation	[130]
Hickory wood	NaOH	600/2	2–100	24	Langmuir	—	53.60	—	[74]
Orange peel	—	500	6–223	6	Langmuir	Second	86.96	Ion exchange and surface precipitation	[49]
Rice straw	KMnO ₄	420/4	1.0 mmol·L ^{−1}	8	Langmuir	Second	305.25	Complexation	[131]
Hickory wood	KMnO ₄	600/1	0–100	24	Redlich Peterson	Richie n-th-order	153.10	Surface adsorption mechanisms	[44]
Swine sludge	Thiourea	300/0.5	0–100	48	Langmuir	Second	145.00	Ion exchange	[80]
Hickory	—	350/5	5–250	24	Dubinin	—	16.30	Cation exchange	[99]
Shell	FeCl ₃ ·6H ₂ O, EDTA	200/8	50–500	12	Langmuir	Second	129.31	Electrostatic interaction and chemical complexation	[110]
Bamboo	—	450/3	2–500	48	Langmuir	First	261.10	—	[132]
Tobacco stem	—	700/2	0–1000	12	Langmuir	—	2047.00	Precipitation	[47]
Bagasse	—	300/2	—	24	Freundlich	Intraparticle diffusion	—	Ion exchange, precipitation	[95]
Sludge	Potassium acetate	700/1	5–300	24	Langmuir	Second	49.47	Complexation, surface precipitation	[133]
Watermelon rind	MgO	600/1	50 mmol·L ^{−1}	24	—	—	742.00	—	[53]
Palm fiber	FeSO ₄ ·7H ₂ O and FeCl ₃ ·6H ₂ O	400/2	25–300	24	Sips model	Second	188.18	Electrostatic interaction, ion exchange, and complexation	[100]
Celery	—	500/3	60–400	24	—	—	304.00	Precipitation, cation exchange, and surface complexation	[134]
Coagulation sludge	MgCl ₂ ·6H ₂ O, MgFe ₃ ·6H ₂ O	500/4	0–140	24	Langmuir	Second	488.78	Ion exchange, electrostatic interaction	[48]

Table 1. Cont.

Feedstock	Modifying Agent	Temperature (°C)/Residence Time (h)	Concentration Range (mg/L)	Contact Time (h)	Isotherm Model	Kinetic Model	Adsorption Capacity (mg/g)	Adsorption Mechanism	Ref.
Pinewood sawdust	Al(NO ₃) ₃ ·9H ₂ O, MgSO ₄ ·7H ₂ O	350/1	10–500	24	Langmuir	Second	591.20	Complexation and electrostatic interaction	[135]
C. Oleifera shells	Polyammonium phosphate	550/1	100–2000	6	Langmuir	Second	723.60	Surface complexation	[81]
Rice straws	β-cyclodextrin	500/4	50–5000	4	Langmuir	Second	131.24	Ion exchange and complexation	[136]
Soybean cake	—	700/2	60	2.5	—	Second	133.60	Cation exchange, precipitation	[137]
Shredded wood	Mild air	475/0.25	0–500	—	—	—	44.00	Precipitation	[60]
Pomelo peel	H ₃ PO ₄	250/2	10–2000	24	Langmuir	Second	88.70	Surface complexation and electrostatic interactions	[52]
Date seed	HCl	550/3	—	—	Sips	—	188.55	—	[38]
Peanut shell	Hydrated manganese oxide	400/1	0–30	24	Freundlich	Second	330.00	Complexation	[96]
Corn stalks	FeSO ₄ ·7H ₂ O, L-cysteine	120/12	10–150	10	Freundlich	Second	103.04	Electrostatic attraction	[97]
Axonopus compressus	Sulfuric acid	180/0.5	0–200	15 min	Langmuir	Second	191.07	Complexation and ion exchange	[70]
Corn stover	ZnO/ZnS	600/1	5–100	48	Freundlich	Second	135.80	Ion exchange, inner sphere complexation	[138]
Eucalyptus globules bark	Zero valent iron	750	0–200	2	Langmuir	Second	60.80	—	[139]
Grape pomace	—	700/2	50–300	24	Langmuir	Second	134.00	Electrostatic attraction, cation exchange, complexation	[51]
Wheat straw	Natural hematite	800/2	5–1500	24	Freundlich	Second	196.91	Precipitation and the surface complexation	[83]
Pine sawdust	Magnetic ferrite	200/8	5–100	6	Langmuir	Second	99.50	Chemical binding adsorption, electrostatic attraction, and ion exchange	[140]
Medulla tetrapanacis	—	700/1	50–400	24	Langmuir	Second	1031.23	Complexation, precipitation, π-π interactions, ion exchange	[46]
Raw sawdust	Magnesium	600/1	20–600	1.5	—	—	202.20	Ion exchange	[141]
Douglas fir	KOH	900–1000/10 s	25–1000	1	Langmuir	Second	140.00	—	[72]
Sludge	Persulfate-zvi	600/1.5	0–100	8	Langmuir	Second	180.00	Electrostatic attraction, electrostatic outer sphere complexation, ion exchange, reduction	[112]
Raw sewage sludge	Potassium hydroxide	350/15 min	10–1000	80 min	—	Second	106.00	Precipitation	[91]

Table 1. Cont.

Feedstock	Modifying Agent	Temperature (°C)/Residence Time (h)	Concentration Range (mg/L)	Contact Time (h)	Isotherm Model	Kinetic Model	Adsorption Capacity (mg/g)	Adsorption Mechanism	Ref.
Quercus robur	FeCl ₃ and FeCl ₂	250/4	25–150	4	Langmuir	Second	63.60	Electrostatic interactions	[117]
Pine wood	MnCl ₂ ·4H ₂ O	600/1	1–300	24	Langmuir and Freundlich	First and second	47.05	Precipitation	[101]
Rice straw	—	550/2	0–1 mmol·L ^{−1}	72	Langmuir	—	0.85 176.12	Chemical complexation	[118]
Rice husk	β-cyclodextrin	300/2	10–300	2	Langmuir	Second	240.13	Electrostatic attraction and complexation	[119]
Water hyacinth	—	433/2.65	0–1000	6	Freundlich	Second	251.39	Precipitation, electrostatic adsorption, surface physical adsorption, ion exchange, and complexation of functional groups.	[121]
Palm oil sludge	—	500/0.5	0–150	4	Freundlich	First, second-	21.76	Boundary layer diffusion	[115]
Cinnamomum camphor	Ultrasound-assisted alkali	450/1	50–1000	6	Langmuir	Second	98.33	Electrostatic attraction and surface complexation	[122]
Fallen leaf	Fe ²⁺ /Fe ³⁺	450/1	0–1000	2	Langmuir	Second	146.84	—	[120]
Bean-worm skin waste	—	500/4	20–300	200	Langmuir	Second	62.00	Chemisorption and precipitation	[123]
Corn stalks	Nanoscale zero-valent iron, KOH	800/2	10–200	8	Langmuir	Second	480.90	Reduction reaction, complexation, and co-precipitation	[124]
Rice husk	Manganese oxide	800/3	15–250	1.5	Langmuir	Second	86.50	Electron density reduction in π-bond aromatic moieties due to the addition of -COOH	[125]
Corn cob-to-xylose residue	Nitrogen doped magnesium oxide	400/2	0–500	24	Freundlich	Second	1429.00	Ion exchange, precipitation and complexation	[42]
Ragweed	—	450/2	1000 ppm	24	Langmuir	Second	358.70	Precipitation, ion exchange, complexation	[142]
Sewage sludge	—	500/0.5	0–320	6	Langmuir	First	41.20	—	[113]
Sludge	—	550/2	200–1000	4	Freundlich	Second	30.88	Co-precipitation, complexation	[128]
Long-root Eichhornia crassipes	—	350/20 min	1.0 mmol·L ^{−1}	50min	Langmuir	Second	118.10	Complexation	[126]
Eucalypts leaf residue	ZnCl ₂ , FeCl ₃ and FeSO ₄	700/2	0–100	48	—	—	52.40	—	[127]
Microcrystalline cellulose	Iron nanoparticles	1000	0–1500	24	Tempkin	Second	17.30	Reduction	[129]
Banana peels	—	600/2	0–600	48	Freundlich	Second	247.10	Electrostatic attraction	[50]

Table 1. Cont.

Feedstock	Modifying Agent	Temperature (°C)/Residence Time (h)	Concentration Range (mg/L)	Contact Time (h)	Isotherm Model	Kinetic Model	Adsorption Capacity (mg/g)	Adsorption Mechanism	Ref.
Rice husk	—	700/1	0–600	48	Freundlich	Second	26.70	Precipitation	[130]
Hickory wood	NaOH	600/2	2–100	24	Langmuir	—	53.60	—	[74]
Orange peel	—	500	6–223	6	Langmuir	Second	86.96	Ion exchange and surface precipitation	[49]
Rice straw	KMnO ₄	420/4	1.0 mmol·L ^{−1}	8	Langmuir	Second	305.25	Complexation	[131]
Hickory wood	KMnO ₄	600/1	0–100	24	Redlich Peterson	Richie n-th-order	153.10	Surface adsorption mechanisms	[44]
Swine sludge	Thiourea	300/0.5	0–100	48	Langmuir	Second	145.00	Ion exchange	[80]
Hickory	—	350/5	5–250	24	Dubinin–Radushkevich	—	16.30	Cation exchange	[99]
Maple wood	H ₂ O ₂	550/	5–550	24	Langmuir	—	43.30	—	[143]
Coconut fiber	—	500/4	50–500	24	Langmuir	Second	175.40	Cation exchange, complexation with functional group, precipitation	[144]
British broadleaf hardwood	—	600	—	24	Langmuir	Second	47.66	Cation exchange	[76]
Raw bagasse	KMnO ₄	600/8	5–200	48	Langmuir	Second	37.45	Precipitation, ion exchange	[145]
Ganoderma lucidum substrate	—	650/2	0–300	24	Freundlich	Second	262.76	Precipitation	[146]
Aerobic granular sludge	FeCl ₃ ·6H ₂ O, FeSO ₄ ·7H ₂ O	200/8	5–150	12	Langmuir	Second	127.00	Surface complexation, electrostatic attraction, and precipitation	[147]
Camellia seed husk	—	700/1	0–300	48	Langmuir	Second	109.67	Ion exchange, complexation, Pb–π interaction, and precipitation	[148]
Mulberry wood	—	650/4	—	—	Freundlich	—	250.00	Ion exchange and chemical precipitation, Pb ²⁺ -p-electrons interaction	[149]
Biogas residue	FeCl ₃ , FeSO ₄ ·7H ₂ O	700/2	25–300	4	Langmuir	Second	181.82	Surface complexation and precipitation, electrostatic attraction	[150]
Printing leaflets	—	600/2	20–400	24	Langmuir	Second	1555.00	Electrostatic interactions, and pi-pi interactions	[151]

Note: — means no relevant data.

5.1. Precipitation

Chemical precipitation is widely used for heavy metal removal in industry, and it can be used effectively in a large temperature range with low operating costs. A large number of studies have shown that precipitation is one of the main Pb removal mechanisms [92,115,122,131]. For example, the surface of biochar had abundant Pb particles, which proved that precipitation was the removal mechanism [114]. The high removal capacity was mainly due to the formation of carbonate mineral precipitation [146]. The peak of CO_3^{2-} shifted after the adsorption of Pb, and the production of the mineral PbCO_3 indicated that Pb(II) was precipitated together with carbonate [152,153]. Besides, phosphates on the surface of biochar also played an important role in promoting the precipitation of Pb. Precipitation of Pb phosphate was an important mechanism for biochar to adsorb Pb(II) [81]. For instance, $\text{Pb}_5(\text{PO}_4)_3\text{Cl}$ [86], $\text{Pb}_5(\text{PO}_4)_3\text{OH}$, and $\text{Pb}_3(\text{PO}_4)_2$ [66] on the biochar proved that Pb phosphate precipitation was an important mechanism.

Furthermore, pH was an important factor affecting the precipitation of Pb ions. $\text{Pb}(\text{OH})_2$ precipitation formed at $\text{pH} > 6.0$, because there were local sites with high alkalinity on the surface of biochar [63,130]. The role of mineral precipitation increased significantly with the increase in pyrolysis temperature [154]. Precipitation of Pb^{2+} and carbonate minerals has been proved to be the main interaction of Pb removal [155]. Some quantitative analyses had also confirmed the predominant role of the precipitation mechanism. For instance, Wu et al. and Cheng et al. found that the contribution of precipitation accounted for more than 50% of the Pb adsorption capacity, because Pb could react with anions (e.g., CO_3^{2-} , PO_4^{3-} , and OH^-) released from biochar to form mineral precipitates [40,156].

5.2. Surface Complexation

Complexation is a process in which electrons interact with donors and acceptors to form various complexes. Complexation plays an important role in the Pb adsorption by biochar. The peak areas of C-O decreased from 31.29% to 21.95% after the adsorption of Pb, which represented that the C-O group combined with a large amount of Pb in solution through surface complexation during the adsorption process [156]. For example, the contribution of surface complexation in the removal mechanism accounted for 55.1% when Gao et al. studied the sorption mechanisms of Pb by rape straw biochar [157]. The content and types of oxygen-containing functional groups on the surface of biochar have a great relationship with the adsorption capacity. Characterization analyses such as X-ray photoelectron spectroscopy and X-ray diffraction showed that the removal mechanism of Pb by biochar was mainly the chemical complexation between Pb and oxygen-containing functional groups. The single bond O functional group might play a dominant role, because the single bond was observed to have a significant decrease in the strength of the $-\text{OH}/\text{C}-\text{O}$ functional group after the Pb adsorption [23]. Compared with the carboxyl group, the hydroxyl group had smaller binding energy and stronger metal complexing ability [118]. Furthermore, the $\text{PbFe}_{12}\text{O}_{19}$ species might be attributed to the complexation between Pb^{2+} and $-\text{OH}$ group on the biochar [158].

Related characterization proved that hydrated manganese oxide nanoparticles could combine with Pb ions through specific inner sphere complexation, and the negatively charged oxygen-containing groups on biochar were beneficial to adsorb Pb [96]. Some studies have shown similar results [126]. Besides, complexation is greatly affected by the preparation temperature of biochar, and the role of surface complexation decreases significantly with the increase in pyrolysis temperature [154].

5.3. Ion Exchange

Ion exchange is the effect or phenomenon of the exchange of ions in a solution with ions on a certain ion exchanger. It uses the exchange of ions in a solid ion exchanger with ions in a dilute solution to remove certain ions in the solution. Ion exchange is also one of the main mechanisms, mainly through ion exchange between the cations on the

oxygen-containing functional groups and the Pb ions. The cation exchange capacity is a vital indicator of the Pb removal effect when the ion exchange is dominant [159].

The oxygen-containing functional group on the surface of biochar is an influencing factor of ion exchange. Besides, the ion exchange mechanism is often influenced by the number of acid sites on the biochar surface [160]. Zhou et al. found that Pb-O/Pb-O=C existed on the surface of banana peels biochar, which proved that the dominant mechanism might be ion exchange [54]. According to the isoelectric point of the carboxyl group and the phenolic hydroxyl group, it was speculated that the main removal mechanism was ion exchange through the carboxyl group [69]. In addition, the adsorption capacity of biochar did not change with the change of specific surface area after desorption, which also proved that the main mechanism was ion exchange [161]. Metal ions such as K^+ and Mg^{2+} , which were equivalent to 10.00% to 38.50% of the total adsorption capacity, were released into the solution, indicating that ion exchange was an important removal mechanism [154].

At the same time, quantitative analysis of some mechanisms also shows that ion exchange plays a major role. Wu et al. found that the contribution of ion exchange accounted for more than 53.31% [40]. The amount of Pb adsorbed by magnesium-modified biochar through mineral precipitation increased by 214.4 mg/g, and the ion exchange adsorption amount of modified biochar was 49 times that of the pristine biochar [40]. Ion exchange is greatly affected by the preparation temperature of biochar, and the role of ion exchange improved significantly with the increase in pyrolysis temperature [154]. In addition, the change of mineral ion exchange sites with pyrolysis temperature is the main factor affecting the adsorption of heavy metal ions [162].

5.4. Electrostatic Interaction

Electrostatic interaction occurs between positively charged Pb ions and negatively charged groups on biochar, especially oxygen-containing functional groups. The density of charges on the surface of biochar attracts ions with opposite charges and drives away ions with the same charge. For example, banana peel biochar had high electronegativity, and electrostatic attraction was the dominant force for the adsorption of Pb on biochar [50]. Besides, Mohubedu et al. found that the electrostatic interaction was the main removal mechanism [117]. Besides, Pb could combine with $-COOH$ and $-OH$ [52], amide oxygen atom, and amino N [163] on biochar by electrostatic interaction. Zeta potential value is an important indicator to explore the removal mechanism of electrostatic attraction. The high electronegativity of the surface of biochar led to the migration of Pb ions to the biochar [50]. Zeta potential was affected by pH, and the adsorption capacity of biochar was poor when $pH < 5.3$. The increase in charge neutralization activity improved the electrostatic interaction when $5.5 < pH < 7$. The adsorption capacity decreased due to the formation of $Pb(OH)_2$ precipitation when $pH > 7.0$. Furthermore, electrostatic interaction of deprotonated functional groups enhanced Pb adsorption [100].

5.5. Chemical Bond

The elements on the biochar can form chemical bonds with the Pb in the aqueous solution to achieve the purpose of removing Pb. The main Pb removal mechanism on biochar involves chemical binding adsorption [140]. The main types of chemical bonds are Pb(II)- π interaction [148] and π - π interactions. π - π interaction played a role in the Pb adsorption by biochar. The π - π interaction promoted the combination of Pb ions and $C=C$ [164]. The shift of $C=C$ and the change of the band after the adsorption of Pb ions indicated that the π - π effect played an important role [46]. Therefore, the deepening of the graphitization degree of biochar favored the adsorption of heavy metal Pb [46]. Besides, the adsorption capacity is consistent with the graphitization degree of biochar, because highly graphitized biochar can provide a large number of π donors [165].

5.6. Physical Adsorption

Physical adsorption also contributes to the adsorption of Pb by biochar. Physical adsorption was a major removal mechanism [46]. In addition, physical adsorption is mainly determined by the specific pore structure and surface area, and physical adsorption is a reversible process [166]. Interaction between biochar and Pb can be achieved through weak van der Waals forces [165]. Gayathri et al. prepared crop waste biochar to adsorb Pb in water [93]. Internal and external diffusion were the main separation mechanism for biochar to adsorb Pb. Besides, pore-filling played an important role in removing Pb [167]. In addition, Lee et al. found that boundary layer diffusion played a major role in the Pb adsorption by biochar [115]. SEM-EDX images showed that the surface of biochar contained Pb element crystal particles, so biochar could adsorb Pb through physical adsorption [121].

6. Problems and Future Perspectives

1. The current studies on the reusability of biochar and the interference of other ions in solution should be strengthened. Meanwhile, the effects of other heavy metals, such as copper and cadmium, on Pb adsorption need to be studied more carefully to facilitate the simultaneous removal of multiple heavy metals.
2. The current research on the Pb adsorption by biochar in wastewater has a large gap with the real Pb polluted water. Thus, the experimental research consistent with the actual polluted water should be promoted.
3. The research content of Pb removal mechanism by biochar remains basically unchanged. The main removal mechanisms include ion exchange, surface complexation, electrostatic adsorption, etc. There is a lack of feedback and improvement processes. Besides, the establishment of analytical models between the preparation conditions and the adsorption capacity might help design experiments more rationally.
4. The morphology and stability of Pb on the biochar surface need to be explored after adsorption, and the Pb leaching in the solution also needs to be studied in detail.

7. Conclusions

1. The results show that different raw materials had different physical and chemical properties. The most popular raw material of biochar for wastewater treatment was agricultural waste, accounting for about 50%.
2. The modification methods of biochar were mainly divided into physical and chemical modification. Among them, the chemical modification was the most widely used and had better effect. At the same time, there were many kinds of modifiers, and the most widely used modifier was metal salt.
3. According to the study of removal mechanism, the results show that the Pb adsorption by biochar mostly conforms to the Langmuir model, and most of the studies on adsorption kinetics are in line with the pseudo-second-order model.
4. Although there were many types of Pb removal mechanisms by biochar in wastewater, precipitation, complexation, ion exchange, and electrostatic attraction are the four main removal mechanisms.

Biochar is a suitable adsorbent for Pb removal in water, and it will be industrially applied widely in the future.

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