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Eco-Friendly Adsorbents for the Removal of Heavy Metals

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Abstract: The continuous discharge of heavy metals has intensified, posing significant risks to ecosystems and human health due to their intrinsic toxicity. Consequently, there is an urgent need to remove these pollutants from wastewater efficiently. Bio-adsorbents, known for their cost-effectiveness and environmentally friendly properties, have become essential tools in water treatment. This review evaluates recent advancements in applying bio-adsorbents for water purification, specifically focusing on understanding the intricate mechanisms governing the adsorption of heavy metals. By identifying key challenges and proposing strategic avenues for future research, the review aims to advance scientific understanding and optimize bio-adsorbents' deployment in advanced water treatment technologies.

Keywords: removal of heavy metals; water treatment; bio-adsorbents; sustainable materials.

1. Introduction

Water, an indispensable resource crucial for the sustenance of living organisms, has assumed heightened importance in the face of contemporary challenges, particularly in developing and emerging nations. These challenges emanate from rapid industrialization, unplanned urbanization, climate change, and the deleterious effects of industrial effluents and domestic waste on water quality [1,2]. The 2018 UN World Water Development Report underscores the alarming statistic that 47% of the global population lacks access to a reliable drinking water supply, a figure predicted to escalate to 57% by 2050, aligning with an estimated global population of 9.4 to 10.2 billion, predominantly concentrated in Africa and Asia [3]. However, the intrinsic value of this vital resource has been significantly compromised since the onset of the Industrial Revolution in the late 18th century, manifesting in the form of water pollution and the consequential onset of chronic and fatal health ailments. The industrial sector, characterized by substantial water consumption, discharges post-processing effluents laden with pollutants into drainage lands or rivers, posing severe threats to aquatic ecosystems. The World Health Organization reports an annual toll of 829,000 deaths attributed to diarrhea resulting from unsafe drinking water, inadequate sanitation, and suboptimal hand hygiene [4]. Wastewater originating from various industrial sectors serves as a reservoir of pollutants, including pesticides, heavy metals, dyes/colors, and a range of hazardous chemicals. As a result, the crucial objective in wastewater treatment processes is thoroughly removing these harmful substances from discharged water.

Among the array of formidable challenges, heavy metals—such as copper (Cu), nickel (Ni), zinc (Zn), cadmium (Cd), arsenic (As), lead (Pb), chromium (Cr), and mercury (Hg)—stand out as prominent threats. These metals insidiously infiltrate the environment through various channels, including applying pesticides, fertilizers, metal-complex dyes, fixing agents (employed to enhance dye absorption onto fibers), mordants, colorants, and bleaching agents [5]. The consequences of sustained exposure to heavy metals are profound, encompassing severe health risks. Notably, the potential for cancer initiation within the human system looms large, driven by the mutagenic processes triggered by these insidious contaminants. Addressing this intricate web of challenges demands a concerted and nuanced approach to safeguarding our most precious resource—water. Common heavy metals, their sources, and associated health concerns are outlined in Table 1.

Various techniques have been employed to extract heavy metals from wastewater, encompassing a range of methodologies such as ion exchange, membrane separation, adsorption, oxidation/reduction, electro-remediation, sedimentation, flocculation, co-precipitation, chemical precipitation complexation, wetland-mediated uptake, photo-catalysis, and, solvent extraction [6–8]. Notably, adsorption, especially leveraging bio-waste materials, has garnered substantial attention due to its economic viability, operational simplicity, and pronounced efficacy in removing metals across a broad spectrum of pH values and from intricate metal forms [6,9]. A gamut of adsorbents, including activated carbon, carbon nanotubes, graphene, and biochar, have been applied, with a proclivity towards bio-waste-derived substrates owing to their operational efficiency, economic viability, and ecologically benign attributes.

Table 1. Maximum allowable limits, origins, and health impacts of hazardous heavy metals [6,12].

Metallic element		Maximum permissible limit (mg/L)	Sources	Health consequences
Arsenic		WHO: 0.01, EPA: 0.05	Extraction processes in mining, metal smelting, and the combustion of fossil fuels	dermatological and carcinogenic effects
Cadmium		WHO: 0.003, EPA: 0.005	Processing of metals, facilities engaged in battery recycling, combustion in power plants, and the emissions from cigarette smoke	Detrimental impacts on the bones, heart, kidneys, lungs, and liver in humans
Chromium		WHO: 0.05, EPA: 0.05 for Cr (VI), 0.1 for Cr(III)	The steel industry and textile manufacturing sector	Impacts on kidney circulation, the development of lung cancer, and the occurrence of dermatitis.
Copper		WHO: 1.0, EPA: 0.25	Mineral extraction and the process of metal smelting	The onset of lung cancer, abdominal pain, diarrhea, liver toxicity, and weakness.
Nickel		WHO: Not specified, EPA: 0.2	Extraction and smelting operations, including mining, as well as industrial sectors such as steel production, automobile manufacturing, battery production, and paint manufacturing.	Renal functioning induces DNA damage, triggers eczema, causes phytotoxicity, and may lead to respiratory cancer.
Zinc		WHO: 3.0, EPA: 1.0	Pharmaceutical production, galvanizing processes, manufacturing of paints, pigments, insecticides, and cosmetics.	Abdominal pain, phytotoxicity, and the develop

The judicious utilization of bio-wastes, exemplified by fruit and vegetable peels, as well as leaves from diverse flora, is accentuated to uphold environmental hygiene. The innovative technology of bio-adsorption involves the fabrication of adsorbents, either in their natural state or chemically modified, derived from botanical sources such as stems, leaves, peels, and husks. This area of research remains vibrant, aiming to offer cost-effective solutions for effectively removing heavy metals from wastewater [10]. The efficacy of the bio-adsorption process is manifest, even under conditions characterized by low concentrations of pollutants in wastewater [11].

This review investigates the potential applications of diverse bio-adsorbents in water purification, specifically highlighting their effectiveness in adsorbing toxic heavy metals. It also highlights significant challenges, promising opportunities, and prospective developments in the realm of bio-adsorbents for water purification. Ultimately, a forward-looking roadmap is proposed, outlining recommendations to guide future research initiatives in this domain.

2. Synthesis

Adsorbents synthesized from raw biomaterials can be manufactured using traditional treatment methodologies. This entails employing established procedures for treating biomaterials to generate the desired adsorbents. Figure 1 illustrates a standardized synthesis process for bio-adsorbents, encompassing the gathering of biomass, cleansing, drying, and size reduction. The drying temperature typically varies between 40 and 120°C, depending on the particular bio-adsorbent [6,13]. In instances where the developed adsorbent fails to achieve optimal efficiency, modifications are imperative before application. Activation of biomass can be accomplished through either high-temperature oxidation during thermal decomposition or low-temperature chemical dehydration reactions [14]. Likewise, producing activated carbon through heat treatment involves two primary steps: pyrolysis and subsequent activation [80]. Biomass pyrolysis is commonly performed below 800°C, and subsequent activation is achieved through chemical or heat treatment approaches. Numerous research studies highlight distinct activation and carbonization temperatures, alongside varied contact durations, in the process of bio-adsorbent synthesis [6,15,16]. The chemical alteration of bio-adsorbents may also be crucial for efficiently eliminating heavy metals [17]. Several research initiatives have investigated various approaches to synthesizing bio-adsorbents. The effectiveness of removing toxic heavy metals from contaminated wastewater is directly influenced by the chosen preparation methods. Common methods for synthesizing bio-adsorbents include physical treatment, chemical treatment, heat treatment, simultaneous heat and chemical treatment, and co-precipitation [6].

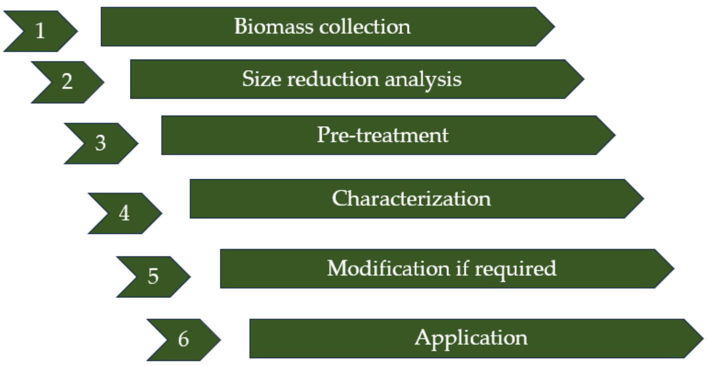

Figure 1. A standardized procedure for the synthesis of bio-adsorbents.

3. Biosorption Mechanism in the Removal of Heavy Metals

Bioadsorbants exhibit an abundance of negatively charged functional groups that are predominantly negatively charged, including hydroxyl, amino, carboxyl, and carbonyl groups. These bio-adsorbents are known for their porous structures, which contain numerous cavities and surface sites that facilitate the binding of metal ions. This porosity significantly increases the surface area available for adsorption, enhancing the bio-adsorbent's capacity to capture and retain metal ions [18]. As a result, the porous nature of bio-adsorbents is essential for their high efficiency and capacity in wastewater treatment applications. The mechanism of heavy metal adsorption from wastewater involves the diffusion of pollutant molecules and their electrostatic attraction to the adsorbent surface. Electrostatic attraction between positively charged metal ions and the negatively charged functional groups on bio-adsorbents is a key factor in increasing adsorption capacity. Hydrophobic interactions, van der Waals forces, and hydrogen bonding can also contribute to the adsorption process on the surface of biosorbents [18]. Complexation and chelation are additional mechanisms that further enhance the adsorption process. For example, the predominant removal of Cr(VI) from aqueous solutions occurs through electrostatic attraction with iron oxide particles on the composite surface of E-BC derived from hickory or bamboo [19]. In ion exchange during adsorption, metal ions substitute the exchangeable ions (Ca^{+2}, Mg^{+2}, Na^{+}) initially present on the bio-adsorbent. The potency of the ion exchange mechanism is influenced by the ionic strength and the accessibility of ionic active sites. A modified biochar obtained from sewage sludge and calcium sulfate demonstrates an ion exchange mechanism for removing Cd(II) from aqueous solutions [20]. Surface complexation involves introducing ligands to the central atom, forming complex ions with metals, and facilitating adsorption. This mechanism, characterized by multiple ligands attracting multiple ions from the solution, is preferred over monoligand complexes. Surface complexation has been identified as an important mechanism in certain biosorption processes [18].

4. Adsorption Application

A thorough investigation of diverse adsorbents obtained from various bio-materials has been conducted to achieve the efficient removal of heavy metals. The superior remediation capabilities of bio-adsorbents arise from their high porosity and the abundance of functional groups. Intrinsic factors such as structure, cation exchange capacity, pore volume, and types of functional groups are pivotal for successfully removing heavy metals with bio-waste-based adsorbents. Operational parameters, including bio-adsorbent dosage, wastewater pH, temperature, coexisting cations, initial concentration, and the specific heavy metals targeted, are crucial determinants in the adsorption process [6,12]. Table 2 displays the efficiencies of various bio-adsorbents in removing heavy metals. in the realm of heavy metal removal from aqueous solutions, researchers have explored many innovative approaches and materials. Wen *et al*. utilized the heteropolysaccharide Ep extracted from *E. prolifera* to synthesize a double-network hydrogel through an interpenetration strategy, showcasing robust mechanical properties and a remarkable adsorption capacity for heavy metal ions (HMIs) [21]. Under conditions where the initial concentration of metal ions is 1600 mg/L, the adsorption capacity of HPEA4 for Pb^{2+}, Cu^{2+}, and Cd^{2+} at room temperature reaches 166.7, 83.3, and 76.9 mg/g, respectively. Similarly, Tan *et al*. investigated cellulose/chitosan aerogels for Cr(VI) adsorption, revealing superior efficacy influenced by chitosan content and pH. With a theoretical Langmuir maximum uptake of 210.6–211.4 mg/g, these aerogels outperformed other materials, as confirmed by Langmuir and pseudo-second-order models. Impressively, the aerogels exhibited durable performance, retaining a Cr-absorption capacity above 75% over six cycles, positioning them as promising water treatment adsorbent [22]. Baby *et al*. presented PKS-Sulfo as a promising adsorbent, demonstrating remarkable efficacy in removing Cr^{6+}, Pb^{2+}, Cd^{2+}, and Zn^{2+} under optimized conditions [23]. The adsorption efficiency was noteworthy, with a 99% removal for Cr^{6+} and Pb^{2+} and a substantial 80-70% removal for Cd^{2+} and Zn^{2+}, respectively. The study highlighted comparable efficiency to activated carbon, with kinetics following the pseudo-second-order model and adherence to Langmuir adsorption isotherm models. In their study, El Mouden *et al*. [24]reported impressive results for the synthesized DS@APTES. The biosorbent exhibited substantial removal efficiencies of 95.29%, 72.35%, and 67.64% for Zn(II), Cu(II), and Pb(II), respectively. The adsorption process was found to conform to both Langmuir and Freundlich isotherm models, highlighting the effectiveness and versatility of DS@APTES. The pseudo-second-order kinetic model also aptly described the adsorption process, emphasizing the chemisorption nature of the interactions between the biosorbent and heavy metal ions. Al-Mahbashi *et al*. [25] explored the effectiveness of sewage sludge-based activated carbon for removing copper and cadmium ions from aqueous solutions. The study employed chemical activation and sulfurization techniques. Through optimization using Box-Behnken Design, the researchers identified optimal conditions resulting in high removal efficiencies of 83.9% for copper and 87.5% for cadmium. This research provides valuable insights into the potential of sewage sludge-based activated carbon as an effective and environmentally friendly solution for heavy metal removal. In their research, A.K. Priya *et al*. [26] investigated the adsorption dynamics of Cr, Pb, and Zn metal ions in an aqueous medium using rice husk ash. The study revealed that optimal adsorption efficiency was achieved under specific conditions: a pH of 6.0, a contact duration of 1 hour, a rice husk dosage of 2.5 g/L, and a temperature of 30°C for 25 mg/L solutions of the targeted metal ions. The rice husk powder demonstrated noteworthy removal capabilities, with removal percentages reaching 87.12% for Cr, 88.63% for Pb, and an impressive 99.28% for Zn. Furthermore, the Temkin and Dubinin-Radushkevich (D-R) isotherm models accurately described the adsorption process, providing a comprehensive understanding of the underlying mechanisms governing the adsorption phenomenon. Similarly, Ahmadi *et al*. introduced Melon Peel as a novel and cost-effective biosorbent for the efficient removal of heavy metals (Cu, Cd, and Pb) from aqueous solutions [27]. Under specified conditions, NMP demonstrated impressive maximum single metal biosorption capacities (q) of 77.76, 76.16, and 191.93 mg/g for Cu, Cd, and Pb, respectively. Furthermore, biosorption kinetics aligned well with the pseudo-second-order model, revealing equilibrium rate constants (k) of 0.07, 0.05, and 0.01 g mg/ min for Cu, Cd, and Pb loaded with NMP. The study also delved into the bio-sorbent's bioavailability, regeneration potential, and reusability, providing a comprehensive understanding of its practical applicability in heavy metal removal scenarios.

In their recent study, Bahman Nazari *et al*. [28] explored the application potential of Pistacia soft shell (PSS) as a biosorbent for the removal of Cu^{2+} and Pb^{2+} ions from aqueous solutions. They achieved notable removal efficiencies of 97% and 98% for Cu^{2+} and Pb^{2+}, respectively, under optimized conditions using 2 g/L of PSS at pH 9. Their mechanistic investigation revealed that the superior performance of PSS can be attributed to its dual-mode action involving both flocculation and adsorption mechanisms, specifically through bridging and sweeping processes. Rahman *et al*. [29] systematically investigated the efficacy of modified shrimp shell chitosan (MSS) as an adsorbent for heavy metal ions in aqueous solutions. Their findings highlighted remarkable removal efficiencies: 98.50% for arsenic, 97.40% (pH 3) for chromium, 74.50% for nickel, and 47.82% for cobalt. The study further determined maximum adsorption capacities of 15.92 mg/g for arsenic, 20.37 mg/g for chromium, 7.00 mg/g for nickel, and 6.27 mg/g for cobalt, corroborated by comprehensive FTIR and SEM-EDS analyses. These results underscore MSS's potential as an advanced and environmentally friendly adsorbent suitable for industrial-scale applications in wastewater treatment.

Table 2. Efficiencies of some bio-adsorbents in removing heavy metals.

Components	Heavy metals	Adsorption condition	Adsorption capacity	Isotherms/ kinetics	References
Enteromorpha polysaccharides-based hydrogels	Pb^{2+}, Cu^{2+}, Cd^{2+}	C= 1600 mg/L,	166.7, 83.3, and 76.9 mg/g, respectively.	Langmuir model	[21]
chitosan and pineapple leaf-based cellulose	Cr(VI)	pH=3, T (◦C)=25 , Adsorbent dosage (g/L)=1	210.6–211.4 mg/g),	Langmuir model	[22]
Palm Kernel Shell	Cr^{6+}, Pb^{2+}, Cd^{2+}, and Zn^{2+}	pH =5, Adsorbent dosage (g/L)=0.25g/L	99% for Cr^{6+} and Pb^{2+} and Cd^{2+} and Zn^{2+} was found to be 80 % and 70%, respectively.	Langmuir isotherm	[23]
Date Stone	Zn (II), Cu (II) and Pb (II)	T (◦C) =25°C, pH 5.5, Concentration = 10 mg/ L, contact time=120min, adsorbent, dosage = 1 g /L	95.29%, 72.35%, and 67.64%	Langmuir and Freundlich isotherms models/ pseudo-second-order kinetic model	[24]
sewage sludge	(Cu) and (Cd)	Cu/ Time=300 min, initial concentration=100 ppm, Adsorbent dose= 20 g/L and pH 6 Initial concentration=11 ppm, 300 minutes contact time, pH 6, and adsorbent dose= 2.5 g/L	83.9% and 87.5% respectively	-	[25]
rice husk powder	Cr, Pb & Zn	pH=6.0, Time=1 h, the rice husk dosage is 2.5 g/L, and temperature of 30°C for 25 mg/L	87.12 %, 88.63 % and 99.28 %, respectively,	Temkin & D-R isotherm model.	[26]
Melon Peel	Cu (II), Cd (II), and Pb (II)	Adsorbent dose= 1.5mg/l pH 6,6, and 7.	77.76, 76.16, and 191.93 mg/g,	Langmuir isotherm/ pseudo-second-order kinetic model	[27]
Orange Peel Cellulose	Cr^{6+}, Cd^{2+}, and Pb^{2+}	Time=36h pH=7	98.33, 93.91, and 33.50, respectively		[30]

5. Conclusion and Future Challenges

In summary, bio-adsorbents, owing to their abundance, cost-effectiveness, and ease of use, have emerged as promising materials for efficiently removing toxic metal ions from water. This review has highlighted recent advances in utilizing bio-waste-derived adsorbents, employing various physicochemical treatments to enhance their sorption capabilities. The adsorption mechanism involves a multifaceted interplay of electrostatic interaction, ion exchange, precipitation, complexation, chelation, and redox processes. The demonstrated effectiveness of bio-adsorbents in heavy metal removal underscores their economic and environmental advantages. Further research is warranted to optimize and broaden bio-adsorbents' application, fostering sustainable solutions for water purification challenges.

Bio-adsorbents present a sustainable and cost-effective solution for treating heavy metal-contaminated water. To fully harness their potential, it is essential to explore their commercial-scale applications, assess environmental toxicity, and innovate for enhanced stability and cost efficiency. Increasing the utilization of industrial bio-waste materials can improve efficacy while minimizing environmental impact. Overcoming challenges such as scaling up from laboratory research to commercial processes, addressing economic constraints, and reducing chemical usage is crucial. Future research should focus on understanding the long-term effects of bio-adsorbents on heavy metals, optimizing the selection of agricultural waste for efficient adsorption, and exploring hybrid bio-adsorbents. Additionally, efforts should target issues like incomplete pollutant removal, high operational costs, energy efficiency, and bio-adsorbent regeneration. Addressing these challenges will drive advancements in water treatment technologies, maximizing the effectiveness of bio-adsorbents.

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Conflicts of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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