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Recent Developments in the Application of Sawdust as a Biosorbent for Heavy Metal Cations: A Mini-Review

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Abstract: Over the last three decades, researchers have recognized the potential benefits of converting sawdust into high-value-added products. Recent advances have been focused on enhancement together with the development of new sawdust biosorbents. This work looks at these current developments, which, through chemical treatment, have increased the sorption capacity for Cu (II) ions, extended the range of operational pH, improved heavy metal recovery, increased biosorbent reuse, and attained WHO drinking water limits. The reviewed literature revealed that these modifications accompanied an increase in biosorbent surface area and pore volume and increased reactivity with the target absorbate. The review also explores the influence of operational parameters, kinetics, equilibrium modeling, and the mechanisms that impact biosorption. In most reported cases, the pseudo-second-order and Langmuir equations provided the best simulation of the biosorption data, forming the basis for rate, monolayer coverage, and biosorption capacity comparisons. The current advances in developing composite biosorbents through functionalization, magnetization, and other chemical treatments have fueled this surge in research interest in using this cheap, easily available material as a precursor. Finally, gaps in our understanding and progress toward the full applicability of sawdust-based biosorbents are identified, and suggestions for future directions are presented.

Keywords: biosorption ; sawdust ; heavy metal ; kinetics ; equilibrium ; biosorption mechanisms.

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1. Introduction

Due to increasing industrialization and technological advancements, there has been a major increase in the discharge of heavy metals into water bodies [1]. The presence of such harmful pollutants at concentrations much greater than the limits set by the World Health Organization (WHO) has raised serious concerns across the globe. According to [2] Qasem *et al*. [2], due to their inability to biodegrade, heavy metals like lead, zinc, mercury, nickel, cadmium, copper, chromium, and arsenic build up in the environment and in living organisms where they can cause a variety of illnesses, interfere with various organ's regular functions, and even be carcinogenic. Copper, for instance, is required in extremely small amounts to preserve a person's health [3]. However, too much copper can result in headaches, diarrhea, breathing problems, liver and kidney failure, and cancer [4]. At the same time, copper is indispensable in the electrical, metallurgical, mining, and agricultural industries because of its qualities, such as strong electrical and thermal conductivity and good corrosion resistance [5,6]. The recommended limit for copper in drinking water is 2 mg/L, per WHO guidelines [6,7].

Thus, it is crucial to remove these harmful heavy metal ions from wastewater before discharge [8].

Many technologies have been thoroughly investigated for removing heavy metals, including ion exchange, filtration, membrane processes, chemical precipitation, and adsorption utilizing activated carbon [9,10]. Unfortunately, despite being successful in metal removal, these techniques often have high capital expenditures or operating expenses and are ineffective at eliminating metal ions in small concentrations [11,12]. As an alternative to expensive traditional methods, biosorption, which uses natural materials, including waste from industrial and agricultural activities, has gained a lot of attention [13-15]. Agricultural waste such as sawdust [16,17], papaya wood [18], sugarcane bagasse [19], neem bark [20], modified pine cone powder [21], coconut husk [22], banana floret [10], black rice husk ash [23], coffee waste [24] and watermelon seed hulls [25], have been evaluated for metal ion sorption. According to Putra *et al*. [5], using agricultural wastes as biosorbents to remove heavy metals has many advantages, including plentifulness, renewables, cheapness, and environmental advantages.

Among the reported agricultural wastes, sawdust is one of the most promising biosorbents for removing impurities such as colors and heavy metals from water and wastewater [26]. In the first quarter of 2023, there has been a surge of research interest in the use of sawdust biosorbents, highlighted in this paragraph. The removal of dye from aqueous solution using sawdust has been recently reported by authors such as Kameni *et al*. [27], where plasma-modified sawdust was used to remove anionic dye. Mallakpour *et al*. [28] prepared an alginate bead embedded with citric-acid-treated sawdust and a metal-organic framework (Zr-based) for the sorption of cationic and anionic dyes. Ahamad *et al*. [29] modified Azadirachta indica sawdust by alkali treatment to produce a sorbent that removed methylene blue almost twice the time of untreated sawdust. Bouyahia *et al*. [30] investigated the removal of methylene blue by cedar wood as a function of extraction techniques viz. maceration, hydrodistillation, ultrasound, and soxhlet. Many recent studies have also focused on using sawdust to prepare activated carbon. Kalak *et al*. [31] used activated carbon from nitrogen-enriched pine sawdust to remove Cu (II) ions. Ilavarasan *et al*. [32] successfully sorbed Cu (II) ions using activated sawdust prepared by heating in a muffle furnace at 200 °C for 24 hours. Nascimento *et al*. [33] prepared a magnetic activated carbon for brilliant blue removal using sapelli sawdust through a single-step pyrolysis method involving KOH and NiCl2. Rzig *et al*. [34] achieved 99.5% removal of tartrazine dye from solution using activated sawdust. Pimentel *et al*. [35] reported on the successful removal of acid dye by activated carbon prepared by chemical activation of pine. Andrade *et al*. [36] prepared hydrochar sawdust by hydrocarbonization between 200 and 240 °C coupled with acid-base treatment. The sorbent successfully removed methylene blue and tartrazine yellow. El-Nemr *et al*. [37] removed acid yellow dye using prepared activated carbon-based sawdust by combining sawdust, urea, fish waste, and zinc chloride, followed by hydrothermal and pyrolysis processes. Drljača *et al*. [38] prepared a carbonized sawdust sorbent for the removal of anionic dye by heating sawdust at 800 °C for 30 min in the absence of oxygen, resulting in more than twice the sorption capacity achieved over that of raw sawdust. Several other sawdust modifications of varying degrees have been recently reported. Chakraborty and Das [39] prepared a sawdust-based nanoparticle by coating sawdust biochar with silica nanoparticles to remove Cr (VI). Gupta and Mondal [40] investigated using biochar derived from sagwan sawdust to remove Cr (VI). Zhou *et al*. 2023 [41] removed Pb (II), Zn (II), and Cu (II) from the solution using a modified fir by microwave-assisted chemical cross-linking. Orozco *et al*. 2023 [42] recently demonstrated the reusability of pine for up to four Cu (II) sorption-desorption cycles with a 94.5% efficiency.

Sawdust mainly comprises organic compounds such as hemicellulose, cellulose, and lignin, as well as hydroxyl, methyl, carboxyl, and amide functional groups in the structures. Both organic substances and functional groups can combine with metal cations and bind them. Furthermore, components like K, Na, and Ca that are part of the timber structure may be crucial for ion-exchange mechanisms [5,26]. Biosorbents in their natural form often exhibit low biosorption capacity and inconstant physical stability [1]. Consequently, research into modifying the biomass surface to achieve structural durability and enhanced biosorption capacity continues unabated. Methods of sorbent modification reported in the literature include physical pre-treatment, such as heating and autoclaving, and chemical treatments using alkaline solutions, organic acid solutions, and organic compounds. In these cases, the improved sorption is generally attributable to the altered surface functional groups, either by masking or eliminating the groups, exposing more metal binding sites, or adding advantageous functional groups [1]. In some instances, acid treatment has increased surface area by converting macropores to micropores. According to several researchers, the magnetization of biosorbents can also increase their surface area, sorption capacity, and efficiency [43,44]. Various biosorbents have also been successfully modified by grafting monomeric units like ethylenediamine, acrylic acid, acrylamide, hydroxylamine, and acrylonitrile. In these instances, these improved biosorption performances were explained by the presence of increased carboxyl groups on the biomass surface.

This paper aims to review current developments in the application and the modifications to sawdust as a biosorbent and assess its impact on heavy metal sorption using copper as a model cation. The reported biosorption capacities, operational parameters that appreciably influence application, modeling, and, where reported, the mechanisms that influence the biosorption process are discussed.

2. Sawdust Biosorption of Cu (II) Ions

2.1. Unmodified coconut tree sawdust.

Putra *et al*. [5] reported on the biosorption of Cu (II) by unmodified coconut tree sawdust (CTS). The biosorbent was prepared by initial washing followed by drying in an oven for 7 days at 70℃. The material was then ground and sieved to 0.15 – 0.25 mm. The surface area (0.4 m2/g) and pore diameter (1.9 nm) were determined by the BET and the BJH methods, respectively. Reaction mixtures were prepared using a biosorbent dose of 4.0 mg/L, pH ranging from 2.0 to 6.0, and initial concentration from 10 – 200 mg/L. The optimum pH was found to be 6.0. The authors revealed that at lower pH, biosorption was inhibited by the presence of H3O+ ions and the protonation of functional groups, while at pH above 6.0, precipitation occurred. SEM analysis before biosorption revealed CTS to be of a smooth and dense surface texture. After biosorption, the authors reported that interactions with Cu (II) ions and the biosorbent resulted in lumps and twist-like deposits on the surface. EDX analysis revealed the surface of CTS, which is composed of carbon and oxygen constituents, and was also able to confirm the biosorption of Cu (II) ions. The presence of functional groups on the surface of CTS was confirmed through FTIR analysis. The authors opined that the interactions with Cu (II) ion and the surface resulted in changes in absorption intensity, the formation of new absorption bands, and shifts in the wavenumber of functional groups. It was confirmed that the surface of CTS comprised amine, hydroxyl, and carbonate as the main biosorption sites. The authors concluded the mechanism of biosorption was attributed to electrostatic attraction (between Cu (II) ions and carbonate group) or complexation (electron pair sharing between O and N). The biosorbent exhibited a strong affinity for Cu (II) ions, which were attributed to Cu (II) high electronegativity (1.93) and ionic radius (0.072 nm). Equilibrium data expressed by the Langmuir isotherm revealed a maximum sorption capacity of 3.89 mg/g.

2.2. Thiol-functionalized magnetic poplar γ-Fe_2O_3 sawdust composites.

Gan *et al.* [8] reported Cu (II) sorption by a thiol-functionalized magnetic sawdust composite. The authors explained that untreated agricultural waste, such as sawdust, can be difficult to separate after biosorption. Further, they can increase water's chemical oxygen demand by releasing pectin, tannin, and lignin. The thiol-functionalized magnetic sawdust composite was developed to overcome these limitations. The sorbent was synthesized by precipitating γ-Fe_2O_3 nanoparticles on poplar sawdust and then modifying 3-mercaptopropyltrimethoxysilane layers. The preparation of the biosorbent involved first washing the sawdust with distilled water, then drying, cutting, and sieving through a 100-mesh screen. A Fe^{3+}/ Fe^{2+} ratio of 2.0 was used to prepare the magnetic sorbent. Wood sawdust (10 g) was added to 200 mL distilled water, followed by 5.40 g $FeCl_3.6H_2O$ (0.02 mol) and 1.98 g $FeCl_2.4H_2O$ (0.01 mol). This was followed by autoclaving at 90 °C to precipitate the magnetic Fe_3O_4 onto the sawdust, then exposing the Fe_3O_4 nanoparticles to air, which led to γ-Fe_2O_3 formation. Finally, the specimens were rinsed and dried at 50 °C for 24 hours. Magnetic sawdust (10 g) was then immersed into the 3-mercaptopropyltrimethoxysilane ethanol solution comprising anhydrous ethanol (100 mL), 3-mercaptopropyltrimethoxysilane (5 mL), glacial acetic acid (5 mL), and H_2O (2 mL) and maintained at 60 °C for 12 hours. Finally, the product was separated, washed, and dried at 50 °C in a vacuum oven. XPS spectra successfully confirmed the modification of the magnetic sawdust with mercaptopropyl silica coating. At the same time, the saturation magnetization was later determined to be 7.28 emu/g and strong enough for separation in 1 min. Adsorption kinetics were best modeled by the pseudo-second-order equation. Equilibrium studies were best modeled by the Langmuir isotherm and revealed a maximum sorption capacity of 5.49 mg/g after 20 min at a pH of 6.0. The authors investigated the desorption and regeneration potential using Pb (II) ions and a 1 M HCl solution under sonication for 30 min. After four successive sorption–desorption cycles, Pb (II) removal was reduced to an efficiency of only 87%.

2.3. Amino-functionalized magnetic poplar γ-Fe_2O_3 sawdust composites.

Gan *et al.* [45] reported on the development of amino-functionalized magnetic γ-Fe_2O_3/sawdust composites (MSC-NH2) for the removal of Cu (II) from solution. The composites were prepared by loading γ-Fe_2O_3 nanoparticles onto sawdust and then treating them with 1,6-hexanediamine. A Fe^{3+}/ Fe^{2+} ratio of 2.0 was used to prepare the magnetic sorbent. Wood sawdust (10 g) was added to 200 mL distilled water, followed by 5.40 g $FeCl_3.6H_2O$ (0.02 mol) and 1.98 g $FeCl_2.4H_2O$ (0.01 mol). This was followed by autoclaving at 90 °C for 8 hours, and finally, the specimens were rinsed and dried at 50 °C for 24 hours. Magnetic sawdust was then added to a solution of 1,6-Hexanediamine (6.5 g) dissolved in ethylene glycol (30 mL) and autoclaved at 110 °C for 8 hours. Finally, the product was separated, washed, and dried in a vacuum oven at 50 °C. Batch kinetics revealed the time to reach equilibrium was 150 min. The pseudo-second-order equation best represented the kinetics, while the Langmuir isotherm produced a maximum sorption capacity of 7.55 mg/g at pH 6.0. Thermodynamic analysis revealed the reaction to be spontaneous, endothermic, and chemical. Desorption of Cu (II) was performed by using 0.1 g of Cu (II)-loaded sorbent and sonicating for 60 min in 100 mL of HCl (0.1 mol/L) solution. This was followed by separation using an external magnet. Consecutive sorption–desorption cycles were carried out five times, reducing efficiency from 97.0% to 78.4 %.

2.4. Citric acid-coated magnetic olive wood sawdust.

The removal of Cu (II) ions by three sawdust biosorbents, viz. citric acid-coated magnetic olive wood (Cit-MOW), magnetic olive wood (MOW), and olive wood (OW) and, was studied by El-Sheikh *et al*. [46]. The authors aimed to develop a biosorbent coated with magnetite, which harnesses magnetic solid phase extraction. Additionally, chemical treatment was employed to modify the functional groups to increase the sorption capacity and the sorbent's durability and rigidity, thus improving regeneration. Olive tree wood was dried and ground to a maximum size of 1.0 mm. The olive wood (150 g) was stirred in 1 L ethanol for 45 min and washed 13 times to remove residual ethanol. To prepare the MOW, 3.36 g of OW was added (mass ratio 1:7) to a mixture comprising distilled water (100 mL), iron (III) chloride hexahydrate (1.16 g), and iron (II) chloride tetrahydrate (0.42 g), and stirred for 1 hour. Finally, 5 ml of ammonia (25%) was added dropwise and stirred for 5 hours in a water bath at 50 °C. Cit-MOW was prepared by stirring 2.5 g of MOW with 25 ml of 0.15 M NaOH for 24 hours. After being washed and dried, 2.0 g of the product was added to 0.40 M citric acid solution (14 mL) and stirred for 1 hour at 70°C. The temperature was then increased for 90 min at 130 °C to initiate thermos-chemical esterification. Finally, the sample was washed with 0.1 M NaOH, separated using a magnet, washed with distilled water, and dried. Biosorption reaction mixtures comprised 25 mg of sorbent in 25 mL Cu (II) solution. The temperature was varied from 30 to 50 °C. Maximum sorption capacities determined by the Langmuir isotherm revealed Cit-MOW, MOW, and OW to be 21, 4.3, and 3.5 mg/g, respectively, at a pH of 5.6. The Dubinin-Radushkevich (D–R) model was used to determine the free energy of adsorption, whereby $8 < E < 16$ kJ/mol implied chemical sorption (ion-exchange) and $E < 8$ kJ/mol implied physisorption. The authors reported the reaction mechanism by Cit-MOW was controlled by physisorption. Thermodynamic analysis revealed the reaction to be spontaneous and endothermic. The authors concluded that bonding citric acid to magnetite increased acidic group content, producing a favorable sorption process.

2.5. Modified Indian rosewood sawdust.

Rani *et al*. [47] reported on Cu (II) biosorption by modified Indian rosewood. Three modifications were reported, viz. sawdust boiled with water (SDB), sawdust treated with formaldehyde (SDF), and sawdust treated with sulfuric acid (SDS). The authors endeavored to enhance the biosorption process by removing water-soluble compounds from the biosorbent, polymerizing color-forming compounds, and increasing the sorbent-available surface area. The rosewood sawdust prepared by boiling treatment was first ground and sieved (< 0.30 mm). The material was then boiled for 8 hours in double distilled water. FTIR analysis confirmed that the boiling process successfully removed compounds that impart color. The sawdust was finally washed and dried at 70℃ for 24 hours. SDF was prepared by mixing the sawdust in 1%

formaldehyde solution (1 part sawdust: 5 parts formaldehyde) for 24 hours at 25℃. The prepared adsorbent was then sieved using a 0.3 mm sieve. Finally, the sorbent was washed to remove residual formaldehyde and activated at 70℃ for 24 hours. The authors used FTIR analysis to confirm the formaldehyde reaction with amine functional groups to immobilize color-forming compounds onto the biosorbent. SDS was developed using (2:1 ratio) sun-dried raw sawdust and concentrated H_2SO_4. The specimen was then heated to carbonizeand soaked in 2% $NaHCO_3$ (w/v) solution to remove excess acid. It was dried in an oven at 70℃ for 24 hours and sieved through 0.3 mm mesh. The authors reported that the SDS possessed the partial carbonization of sawdust due to sulfuric acid treatment. Characterization studies revealed the major functional groups in untreated rosewood sawdust, SDB, SDF, and SDS, including C=O, −COOH, −OH, and $-NH_2$. The BET surface area and pore volume were analyzed for SDB (235.786 m^2/g; 0.180 cm^3/g), SDF (285.385 m^2/g; 0.214 cm^3/g) and SDS (514.996 m^2/g; 0.426 cm^3/g). The authors reported that overall, the SDS exhibited the highest percentage removal compared to SDB and SDF, which they attributed to its greater surface area as indicated by BET analysis. The SDB experimental data were most accurately represented by both the Freundlich and Langmuir isotherms. The SDF was best modeled by the Freundlich isotherm. SDS was best simulated by the Langmuir model, which implied uniform monolayer biosorption—at a pH of 5.8, SDB, SDF, and SDS produced exceptionally high biosorption capacities of 98.81, 178.57, and 142.85 mg/g, respectively. The pseudo-second-order equation was the most accurate representation of biosorption kinetics. Thermodynamic analysis for SDS revealed that Cu (II) biosorption was feasible, endothermic, and occurred with increased randomness. Biosorption using SDF and SDB was reported to be exothermic and took place with a decrease in randomness between the solid-liquid interfaces. Desorption studies revealed better removal by acid eluents than NaOH and KI. Using 0.2 N HCl, a desorption efficiency of 69.9 %, 63.82 %, and 84.78 % was achieved for SDB, SDF, and SDS, respectively.

2.6. Modified poplar, cherry, spruce, and hornbeam sawdust.

Kovacova *et al*. [4] investigated the removal of Cu (II) by modified cherry, poplar, hornbeam, and spruce sawdust. By treating the material with sodium hydroxide or potassium hydroxide, the authors showed that residual lignin was eliminated, and new sorption sites were created. Cherry, poplar, hornbeam, and spruce sawdust were first sieved to a maximum particle size of 2.0 mm. Without stirring, 20 g of wooden sawdust was mixed in 200 mL of either a 1 M KOH or 1 M NaOH solution and left for 24 hours. The specimens were separated, washed, and dried in an oven at 45 ± 5 ◦C. Biosorption reaction mixtures comprised 0.5 g sawdust in 50 mL of synthetic copper solution at concentrations ranging from 10 to 150 mg/L at an initial pH of 4.0. According to the authors, treatment with NaOH resulted in increased hydroxyl functional groups for cherry, spruce, and hornbeam sawdust. Meanwhile, treatment with KOH considerably intensified the hydroxyl functional groups only in the poplar sawdust. Biosorption equilibrium studies were best modeled by both the Langmuir and Freundlich isotherms. Langmuir's model revealed maximum sorption capacities for untreated poplar, cherry, spruce, and hornbeam of 3.97, 2.08, 2.13, and 4.32 mg/g, respectively. NaOH-treated poplar, cherry, spruce, and hornbeam sorption capacities were reported as 8.20, 7.38, 9.02, and 7.71 mg/g, respectively. KOH-treated poplar, cherry, spruce, and hornbeam produced sorption capacities of 7.86, 11.06, 10.52, and 9.49 mg/g, respectively. The authors concluded that the major mechanisms involved in Cu (II) removal were complexation and ion exchange.

2.7. Poly(N-acryloyl-L-histidine) modified birch sawdust.

Gao *et al*. [48] studied the design of poly(N-acryloyl-L-histidine)-modified sawdust (SD-g-P(A-His)). The biosorbent was prepared by reversible addition–fragmentation chain transfer (RAFT) polymerization of N-acryloyl-L-histidine on birch sawdust. The authors intended to use birch sawdust as a carrier onto which ploy(N-acryloyl-L-histidine) was grafted, resulting in imidazolyl and carboxyl groups that would chelate Cu (II) ions from solution. This composite biosorbent was designed to target metal ions selectively, reduce metal concentrations below drinking water standards, and improve reusability. Synthesis of N-acryloyl-L-histidine involved first dissolving L-histidine (10.0 g, 73.0 mmol) in NaOH (32 mL, 2.5 M) and acryloyl chloride (6.7 g, 73.6 mmol). Acetone was used to precipitate the unreacted L-histidine after 12 hours of stirring, after which N-acryloyl-L-histidine was recovered. To synthesize the sawdust-based macroinitiator (SD-CTA), a slurry comprising sawdust (10.0 g), methylene chloride (40 mL), and triethylamine (1.0 g, 9.9 mmol) was prepared. After cooling to 10 °C, DDACl (3.4 g, 9.0 mmol) in methylene chloride (2 mL) was mixed with the slurry. The slurry was stirred for 24 hours before being poured into methanol, then washed and dried in a vacuum at 35 °C. The preparation of poly(N-acryloyl-L-histidine) sawdust involved adding 30 mL of N,N-dimethylacetamide to AIBN (19.6 mg; 0.12 mmol) and SD-CTA (10 g, CTA 0.8 mmol). Finally, N-acryloyl-L-histidine (10 g; 47.8 mmol) was added and stirred. Polymerization was performed at 80 °C for 24 hours, then precipitated in methanol to produce SD-g-P(A-His). Biosorption kinetic studies revealed an initial rapid uptake followed by a slow approach to equilibrium which occurred after 60 min. The pseudo-second second-order model well simulated the adsorption kinetics. The maximum reported biosorption capacity of SD-g-P(A-His) for Cu (II) ions was 95.7 mg/g. Untreated birch attained a maximum biosorption capacity of 1.6 mg/g. The authors attributed the outstanding Cu (II) removal to the presence of imidazolyl and carboxyl groups on the biosorbent. Further, they explained that this imparted a pH-responsive biosorption behavior, where specific metal ions can be targeted. The authors studied Pb (II) desorption from SD-g-P(A-His) using HCl and EDTA solution for 1 hour. After five sorption-desorption cycles, the biosorption efficiency dropped to only 91.5 %. The authors concluded that SD-g-P(A-His) could attain low treatment levels of less than 10 µg/L.

2.8. NaOH-modified Michelia figo sawdust.

Long *et al*. [49] investigated Cu (II) biosorption by NaOH-treated M. figo sawdust. The preparation of the biosorbent involved reducing the size between 0.25 and 0.50 mm, followed by washing and drying. The specimen was soaked in NaOH (0.5 mol/L) solution for 4 hours. This was followed by washing with de-ionized water and drying at 45 °C. The biosorption reaction mixture comprised 100 ml of Cu (II) solution agitated at 150 rpm, a temperature of 25 °C, a pH of 5.0, a biosorbent dose of 4 g/L, and an initial metal concentration of 100 mg/L. The authors reported a maximum Cu (II) removal of 14 mg/g. The initial metal ion concentration was reported to be the most significant variable regarding Cu (II) biosorption. The performance of the biosorbent for Cu (II) ions was ascribed to the presence of hydroxyl and carboxyl groups on NaOH-treated M. figo.

2.9. Unmodified Acacia nilotica sawdust.

Aachhera *et al*. [50] reported on Cu (II) biosorption by unmodified *Acacia nilotica* sawdust. The *Acacia nilotica* sawdust was first sieved to a size between 3.0 to 0.03 mm.

Without chemical treatment, the sieved sawdust was used for biosorption experiments. The maximum sorption capacity attained at pH 4.0 and a biosorbent mass of 0.8 g was reported to be 1.85 mg/g. The equilibrium data was best modeled by the Langmuir isotherm, while the pseudo-second second-order model best-simulated kinetics. The authors concluded that the rate-limiting step may have been chemisorption involving polar functional groups on *Acacia nilotica* and covalent forces by sharing or exchanging electrons between these groups and Cu (II) ions.

3. Biosorbent Preparation

The preparation of sawdust biosorbents is categorized here into three stages (Table 1). Stage 1 preparation comprises grinding, sieving, washing, and drying. Washing typically involves using distilled or de-ionized water to remove small dirt and dust particles from the sawdust. To preserve the biosorbent's characteristics, most authors reported drying the sorbent in an oven and maintaining the temperature between 45 to 50 °C for 24 hours. Where reported, grinding devices include mortar and pestle, mill, or electronic grinders. Particle sizes used for experimentation were kept less than 3.0 mm, with most researchers maintaining the range of 0.15 to 0.3 mm. Stage 2 preparation involves physical, chemical, or thermal treatment of the sorbent to enhance the existing properties, free occluded pores, increase specific surface area, increase pore volume, immobilize impurities, and mechanical and other non-biosorptive modifications such as magnetization to improve separation. Rani *et al*. [47] experimented with rosewood, where they successfully induced boiling to remove impurities that impart color, treated with formaldehyde to immobilize the impurities that impart color, and treated with sulfuric acid to increase the surface area and pore volume. Kovacova *et al*. [4] showed that alkali treatment with NaOH or KOH removed residual lignin and created new sorption sites by intensifying hydroxyl function groups on the sawdust surface. Gan *et al*. [8] and El-Sheikh *et al*. [46] successfully prepared magnetic sawdust particles through precipitation using Iron (III) chloride hexahydrate and iron (II) chloride tetrahydrate. Stage 3 preparation involves advanced functionalization processes such as grafting, polymerization, and cross-linking to coat the sorbent to enhance or add biosorption properties. Gan *et al*. [8] successfully added -SH functional groups by mercaptopropyl silica coating on poplar sawdust. Gao *et al*. [48] modified birch sawdust by coupling the hydroxyl group with DDACl and grafting poly(N-acryloyl-L-histidine) via RAFT polymerization. El-Sheikh *et al*. [46] reported on citric acid treatment of olive wood sawdust, which increased total acidic groups, carbon content, and methylene blue surface area. Additionally, there was a minimal decrease in BET surface area and pHpzc, while the total basic group on the biomass remained unchanged.

Table 1. Stages of biosorbent preparation.

Biosorbent	**Stage 1 Preparation**			**Stage 2 Preparation**	**Stage 3 Preparation**	**Comments**	**Ref.**
	Sieving	**Washing**	**Drying**	**Physical and chemical modifications**	**Grafting, polymerization & esterification**		
Thiol-functionalized magnetic poplar γ-Fe_2O_3 sawdust composites	< 0.15 mm	Distilled water	50 ℃ for 24 hours.	Precipitation of Iron (III) chloride hexahydrate & iron (II) chloride tetrahydrate	Addition of –SH functional group by mercaptopropyl silica coating	Modification of magnetic sawdust with mercaptopropyl silica coating accompanied by increased pore volume and surface area	[8]

Biosorbent	**Stage 1 Preparation**			**Stage 2 Preparation**	**Stage 3 Preparation**	**Comments**	**Ref.**
	Sieving	**Washing**	**Drying**	**Physical and chemical modifications**	**Grafting, polymerization & esterification**		
Amino-functionalized magnetic poplar γ-Fe_2O_3 sawdust composites	< 0.15 mm	Distilled water	50 ℃ for 24 hours.	Precipitation of iron (III) chloride hexahydrate & iron (II) chloride tetrahydrate	Addition of amino-functionalized groups using 1,6-hexanediamine	Increased sorption sites due to amino functional groups, which accompanied an increased pore volume and surface area	[45]
Citric acid-coated magnetic olive wood	< 1.0 mm	Ethanol & distilled water	50 ℃ for 24 hours.	Precipitation of iron (III) chloride hexahydrate & iron (II) chloride tetrahydrate followed by alkali treatment	Bonding citric acid to biomass to increase acidic functional groups	Increased in total acidic groups and methylene blue surface area, marginally decreased the BET surface area and pH_{pzc}.	[46]
Rosewood boiled with water (SDB)	< 0.30 mm	Distilled water	70℃ for 24 hours.	Biomass boiled in distilled water for 8 hours.		Boiling treatment to remove the color-imparting impurities	[47]
Rosewood modified with formaldehyde	< 0.30 mm	Distilled water	70℃ for 24 hours.	Biomass soaked in 1% formaldehyde solution at 25℃ for 24 hours followed by activation at 70℃ for 24 hours		Immobilization of the water-soluble colored impurities by polymerization	[47]
Rosewood modified with sulfuric acid	< 0.30 mm	Distilled water	70℃ for 24 hours.	Biomass soaked in concentrated H_2SO_4 followed by carbonizing under heating.		−OH group acidification and an increase in biomass surface area	[47]
NaOH modified poplar	< 2.0 mm	Distilled water	45 ℃	20 g sawdust in 200 mL 1 M NaOH for 24 hours without mixing		Residual lignin removal, increased porosity and the creation of new sorption sites through intensification of hydroxyl functional groups	[4]
KOH modified cherry	< 2.0 mm	Distilled water	45 ℃	20 g sawdust in 200 mL 1 M KOH for 24 hours without mixing		Intensification of hydroxyl functional groups	[4]
KOH modified spruce	< 2.0 mm	Distilled water	45 ℃	20 g sawdust in 200 mL 1 M KOH for 24 hours without mixing		Intensification of hydroxyl functional groups	[4]
KOH modified hornbeam	< 2.0 mm	Distilled water	45 ℃	20 g sawdust in 200 mL 1 M KOH for 24 hours without mixing		Intensification of hydroxyl functional groups	[4]
Poly(N- acryloyl-L-histidine) modified birch sawdust	< 0.84 mm	Ultra-pure water	35 °C for 24 hours.	Modification through coupling the hydroxyl group with DDACl.	Poly(N-acryloyl-L-histidine) grafted onto the sawdust by RAFT polymerization	Formation of imidazolyl and carboxyl groups	[48]
NaOH modified *M. figo*	0.25 - 0.5 mm	De-ionized water	45 ℃	Biomass soaked in 0.5 mol/L NaOH for 4 hours at room temp.		Intensification of hydroxyl and carboxyl groups of NaOH-treated *M. figo*	[49]

4. Elucidating the Performance of Sawdust Biosorption of Cu (II) Ions

Table 2 compares some of the noted performances of sawdust biosorption of Cu (II) ions from the literature and those reported in this review. The table reveals that over the past decade, there has been a great stride in examining and modifying sawdust biosorbents. Attainable treatment levels, reusability, maximum attainable capacities, and pH operational ranges have gained further insight. Gao *et al.* [48] developed a poly(N-acryloyl-L-histidine)-modified birch sawdust which could reduce initial Cu (II) concentration from 1.0 mg/L to less than 10 μg/L, below WHO treatment limits. The biosorbent could also significantly reduce Cu (II) levels within the pH range of 2.7 to 4.7 (Table 3). Gan *et al.* [8] and Goa *et al.* [48] reported that poly(N-acryloyl-L-histidine)-modified birch sawdust and thiol-functionalized magnetic poplar sawdust were able to achieve an 87% (after 4 cycles) and 91% (after 5 cycles) biosorption efficiency after successive sorption/desorption cycles, respectively. Rani *et al.* [47] achieved maximum sorption capacities of 178.6 and 143.0 mg/g for rosewood treated with formaldehyde and rosewood treated with sulfuric acid, respectively. After pre-treatment, the authors attributed this significant removal to a marked improvement in pore volume and specific surface area. Table 3 also presents some key characteristics that influenced the biosorption process. Where reported, most biosorbents followed and were best simulated by the pseudo-second-order and the Langmuir model. Some studies reported the mechanism of biosorption to be influenced by electrostatic attraction between metal ions and functional groups, while most reported complexation involving electron sharing or ion exchange.

Table 2. Maximum biosorption capacity of different sawdust biosorbents for Cu (II) removal.

Biosorbent	**Wood specie**	**q_m (mg/g)**	**Ref.**
Unmodified poplar sawdust	*Populus sp.*	3.2	[51]
HCl-modified Oak sawdust	*Quercus sp.*	3.5	[52]
Formaldehyde and NaOH-modified oak sawdust	*Quercus sp.*	12.4	[53]
Hot-alkali modified teak sawdust	*Tectona sp.*	8.4	[54]
Unmodified beech sawdust	*Fagus sp.*	4.5	[55]
Unmodified birch sawdust	*Betula sp.*	4.9	[56]
Unmodified mansonia sawdust	*Mansonia sp.*	42.4	[57]
Unmodified coconut tree wood	*Cocos sp.*	3.89	[5]
Thiol-functionalized magnetic poplar γ-Fe_2O_3 sawdust composites	*Populus sp.*	5.49	[8]
Amino-functionalized magnetic poplar γ-Fe_2O_3 sawdust composites	*Populus sp.*	7.55	[45]
Unmodified olive wood	*Olea sp.*	3.5	[46]
Magnetic olive wood	*Olea sp.*	4.3	[46]
Citric acid-coated magnetic olive wood	*Olea sp.*	21	[46]
Rosewood boiled with water (SDB)	*Dalbergia sp.*	98.81	[47]
Rosewood modified with formaldehyde	*Dalbergia sp.*	178.57	[47]
Rosewood modified with sulfuric acid	*Dalbergia sp.*	142.85	[47]
NaOH modified poplar	*Populus sp.*	8.2	[4]
KOH modified cherry	*Prunus sp.*	11.06	[4]
KOH modified spruce	*Picea sp.*	10.52	[4]
KOH modified hornbeam	*Carpinus sp.*	9.49	[4]
Unmodified birch wood	*Betula sp.*	1.6	[48]
Poly(N- acryloyl-L-histidine) modified birch sawdust	*Betula sp.*	95.7	[48]
NaOH modified M. figo	*Michelia sp.*	14	[49]
Unmodified *Acacia nilotica*	*Acacia sp.*	1.85	[50]

Authors have shown pH significantly influences Cu (II) ions' biosorption. In all reviewed cases, the pH was kept below 6.0 to prevent precipitation, with optimal conditions being observed between pH 4 and 5.8. This is advantageous as it mitigates to some degree the burden of cost to condition waters before biosorption.

Table 3. Key characteristics influencing biosorption mechanisms.

Biosorbent	pH	Kinetic Model	Equilibrium Model	Mechanism	Ref.
Unmodified coconut tree wood	6.0	-	Langmuir equation	Electrostatic attraction or complexation	[5]
Thiol-functionalized magnetic poplar γ-Fe_2O_3 sawdust composites	6.0	Pseudo-second order equation	Langmuir equation	Chemical sorption	[8]
Amino-functionalized magnetic poplar γ-Fe_2O_3 sawdust composites	6.0	Pseudo-second order equation	Langmuir equation	Chemical sorption	[45]
Unmodified olive wood	-	-	Langmuir equation	-	[46]
Magnetic olive wood	-	-	Langmuir equation	-	[46]
Citric acid-coated magnetic olive wood	-	-	Langmuir equation	Physisorption	[46]
Rosewood boiled with water (SDB)	5.8	Pseudo-second order equation	Freundlich and Langmuir equations	-	[47]
Rosewood modified with formaldehyde	5.8	Pseudo-second order equation	Freundlich equation	-	[47]
Rosewood modified with sulfuric acid	5.8	Pseudo-second order equation	Langmuir equation	-	[47]
NaOH modified poplar	-	-	Freundlich and Langmuir equations	Complexation and ion exchange	[4]
KOH modified cherry	-	-	Freundlich and Langmuir equations	Complexation and ion exchange	[4]
KOH modified spruce	-	-	Freundlich and Langmuir equations	Complexation and ion exchange	[4]
KOH modified hornbeam	-	-	Freundlich and Langmuir equations	Complexation and ion exchange	[4]
Unmodified birch wood	-	Pseudo-second order equation	-	-	[48]
Poly(N- acryloyl-L-histidine) modified birch sawdust	2.7-4.7	Pseudo-second order equation	-	-	[48]
NaOH modified *M. figo*	5.0	-	-	-	[49]
Unmodified *Acacia nilotica*	4.0	Pseudo-second order equation	-	Chemical sorption	[50]

5. Future Prospects and Current Challenges

Despite the strides in laboratory testing of sawdust biosorbents for heavy metal uptake, there are still areas that require significant emphasis in research. The optimization of batch-scale operational parameters, characterization, and mechanisms have been well documented. However, there is little emphasis on column application and resolving issues related to bed uniformity and compressibility, which can lead to increased head loss. Most studies have focused on synthetic single-metal ion solutions; however, more research is needed on real-world scenarios involving multi-metal ion solutions and the impacts of competing and coexisting ions. To realize the full benefit of sawdust biosorption, cost comparisons need to be investigated, chiefly the cost of modifications. Sawdust biosorption has the potential to become an attractive treatment alternative, particularly for small and medium-sized industries that cannot afford the cost of present commercial sorbents.

6. Conclusions

The removal of heavy metals from solutions using sawdust-biosorbent was discussed in this review paper. Copper was selected as a model cation. Considerable progress has been made in the development and use of sawdust for the biosorption of heavy metals from aqueous streams over recent years. Such developments can aid in providing a useful purpose for this natural waste. To improve the performance of sawdust, various chemical treatments have proven to increase the sorption capacity for Cu (II) ions, extend the range of operational pH, improve heavy metal recovery, increase biosorbent reuse, and enable the achievement of WHO drinking water limits. These enhancements in performance have been attributed to an increase in biosorbent surface area, pore volume, and increased reactivity with the target absorbate. Operational parameters such as equilibrium time, solution pH, initial Cu (II) concentration, biosorbent dose, and temperature have been studied to elucidate the optimal biosorption process conditions. In all reported cases, the pseudo-second-order equation produced a good representation of the reaction kinetics. Also, in most reported cases, the Langmuir isotherm provided the best representation of the equilibrium data, forming the basis for monolayer coverage and biosorption capacity comparisons. Some studies reported the influence of electrostatic attraction, while most concluded that complexation involving electron pair sharing or ion exchange was the primary mechanism. The review also revealed gaps in our understanding and progress towards the full applicability of sawdust-based biosorbents. More research is needed to experiment and model packed-bed systems to elucidate the scalability and real-use applicability. Further, little effort has been made to explain the cost between sawdust biosorption, their modifications, and conventional systems. The application of mixed-metal solutions also needs further exploration to elucidate the effects of competing ions in the solution.

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Conflicts of Interest

The authors declare no conflict of interest.

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