

A review of the advancements of potentially toxic element adsorption by various cellulose-based materials and the used adsorbents' fate

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ABSTRACT

In this study, we compile the findings to date on using several cellulose-based materials as adsorbents of potentially toxic elements (PTEs) from wastewater. Furthermore, this review discussed the destiny of PTEs-loaded cellulose-based adsorbents and some sustainable methods for their management, hoping to close the pollution loop.

KEYWORDS

potential toxic elements (PTEs), cellulose, paper, wood mulch, adsorbent fate

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INTRODUCTION

Potentially toxic elements (PTEs) are ubiquitous environmental pollutants that significantly threaten human health because they tend to intoxicate, accumulate in, and remain in ecological media (Nguyen et al., 2020) classified potentially toxic elements as hazardous pollutants, especially in aquatic ecosystems. PTEs pollution is a global environmental issue due to the continuous discharge of pollutants from various sources, including industrial effluents, mining activities, agricultural runoff, and improper waste disposal.

Traditional methods for treating wastewater contaminated with PTEs ions include chemical processes (oxidation and reduction, ion exchange filtration, precipitation, and electrochemical treatments) and other physical processes like evaporation treatments (Volesky, 1994; Vievard et al., 2023). However, as stated by Veglio and Beolchini (1997), these high-technology processes have significant drawbacks, such as incomplete metal removal, expensive equipment and monitoring systems, high reagent energy, and the production of toxic sludge or other waste products that need to be disposed of. Therefore, the development of effective and eco-friendly adsorbents for the removal of PTEs is of paramount importance. Therefore, selecting, developing, and characterizing adsorbent materials is critical in designing an adsorption process for water treatment. Adsorbents for water treatment must have the following characteristics: low cost and availability, chemical stability, mechanical stability, good textural and physicochemical properties, high adsorption capacity, high efficiency, fast kinetics, and the ability to regenerate and reuse (Dotto and McKay, 2020). Many materials, including agriculture products, red mud, clay minerals, fly ash, Portland cement, and cellulose-based materials, have been tested as cheap and abundant adsorbents (Mondal et al., 2019; Nag and Biswas, 2021).

Biological materials, including living and non-living biomaterials, have been widely used to remove pollutants in wastewater, such as PTEs, ions, and dyes (Çolak et al., 2009; Chu and Phang, 2019; Kratochvil and Volesky, 1998). The offered advantages of the biosorption process are the low operating cost, the possibility of regeneration of biosorbents, metal recovery, and the minimization of the quantity of chemical and biological sludge needed to be managed (Kratochvil and Volesky, 1998; Beni and Esmaeili, 2020).

In recent years, cellulose-based adsorbents have attracted much interest as a potentially helpful material for removing PTEs from contaminated water. Cellulose is an attractive material for the adsorption of PTEs due to its unique physicochemical properties, such as its high surface area, biocompatibility, and low cost. Physical and chemical processes, including ion exchange, electrostatic interactions, and complexation, are involved in the adsorption of PTEs onto bio-based adsorbents (Kurniawan et al., 2023). Most cellulose-based adsorbents follow the pseudo-second-order kinetic model, which assumes that the rate-limiting step is chemisorption (Syeda and Yap, 2022). Other researchers modified cellulose-based adsorbents' surface chemistry and morphology through various methods such as chemical modification, physical treatment, and grafting of functional groups to enhance their adsorption capacity and selectivity (de Quadros et al., 2016).

Researchers have investigated the efficacy of many cellulose-based adsorbents in removing PTEs from polluted water. However, the existing data is scattered; a comprehensive study is necessary for discussing the field's past and current advancements. Therefore, this review further discussed some research questions, such as the fate of the cellulose-based adsorbents loaded with PTEs, which is rarely studied in the literature but is a significant concern due to the potential environmental impacts they may cause.



CELLULOSE

Cellulose is nature's most prevalent biopolymer and is the primary component of plant fibers, which provide plant rigidity (Sharma et al., 2019). It is a long-chained linear polysaccharide composed of β -D-glucopyranose units linked by β -1,4 glycosidic linkages (Faruk et al., 2012; Henriksson and Berglund, 2007; O'Connell et al., 2008). Figure 1 shows the chemical structure of cellulose.

Cellulose-based materials have been used in various construction applications, primarily as intact wood. Furthermore, cellulose is abundant in commonly used materials such as cotton (90%), wood (50%), and dried hemp (57%). It has many applications in various fields but is most commonly used in producing paper, cardboard, and derivative products such as cellophane and rayon. It is also a significant component of cotton and linen textiles. In addition, powdered cellulose and microcrystalline cellulose are used in the pharmaceutical industry as inactive drug fillers. Also, cellulose is a versatile starting material for chemical conversions that produce artificial cellulose-based threads and films and several stable cellulose derivatives utilized in various industrial and consumer applications (Gupta et al., 2019).

The cellulose content is variable in agricultural and waste materials; some cellulose content in different materials is presented in Table 1.

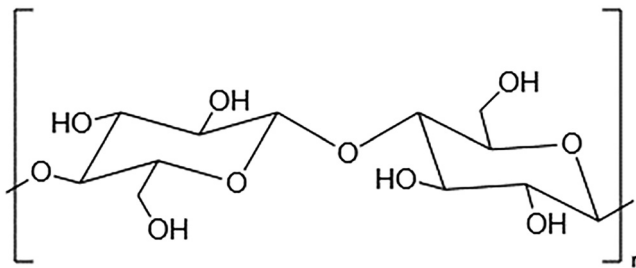


Fig. 1. Chemical structure of cellulose

Table 1. The cellulose content in different materials

Material	Cellulose content (%)	Source
Oakwood	41	Le Floch et al. (2015)
Wastepaper	90–99	Sahin and Arslan (2008)
Cotton	95	Holtzapple (2003)
Leaf fiber	55–73	Hokkanen et al. (2016)
Paulownia wood	43.93	Huo et al. (2022)
Crop residues	30–50	Koul et al. (2022)
Wheat straw	30	Sundarraj and Ranganathan (2018)
Jute fiber	58–63	Wang et al. (2008)
Maize Straw	28–44	Rehman et al. (2014)
Coconut waste	28.7	Rojas et al. (2018)



Several natural cellulose-based materials have been tested by researchers and found promising results for PTEs removal, such as coconut shells (Tan et al., 1993; Low et al., 1995; Baes et al., 1996; Pino et al., 2006); wood sawdust (Sharma and Forster, 1994; Mohan and Singh, 2002; Dakiky et al., 2002; Šćiban and Klačnja, 2003; Acar and Eren, 2006; Gupta and Babu, 2009; Putra et al., 2014), and paper waste as adsorbents for the removal of dyes and PTEs from wastewater (Fahad et al., 2018). These eco-friendly adsorbents show good affinity towards PTEs due to the cellulose content in these materials, which have good adsorption potential due to O-containing and hydroxyl functional groups (Jamshaid et al., 2017). Surface complexation, ion exchange, and electrostatic contact were all mentioned as adsorption methods by which these functional groups may efficiently coordinate with PTEs (Han et al., 2022). Binding affinity is greatly affected by whether PTE ions establish bonds with monodentate or bidentate functional groups (Zhang et al., 2020). In addition, PTEs' charge and valency affect the interface's electrical characteristics. Metal ions with positive charges, such as Cd (II), Pb (II), and Hg (II), have an electrostatic attraction towards functional groups with a negative charge. Arsenate (AsO_4^{3-}) ions, which are negatively charged, prefer to attach to positively charged sites. According to Han et al. (2022), the electrostatic interactions between PTE ions and functional groups are enhanced for divalent and trivalent PTE ions compared to monovalent PTE ions due to the valency effect.

Most adsorption studies were conducted using untreated cellulosic materials, and only a few demonstrated good adsorption potential. Other researchers applied physical and chemical treatment, which significantly impacted the performance of these adsorbents. For example, Šoštaric et al. (2018) applied treatment with NaOH to apricot shells, increasing the adsorption capacity by 90%, 154%, and 61% for Pb^{2+} , Cu^{2+} , and Zn^{2+} , respectively. Wang et al. (2019) developed a carboxymethylated cellulose fiber for water purification. The prepared carboxymethylated cellulose fiber bio-adsorbent removed Cu (II) more effectively than the unmodified fibers, whose adsorption capacity increased 130-fold. In another study, Huo et al. (2022) used a wood-based adsorbent modified by esterification with phosphoric acid. The ideal adsorption conditions were 318.15 K with $\text{pH} = 6$, and the phosphorylated wood maximum adsorption capacity was 130.2 mg g^{-1} , seven times higher than that of the alkaline extracted wood (18.5 mg g^{-1}).

The most recent studies are concentrated on synthesizing cellulose- and nano-cellulose-based adsorbents. For example, maleic acid-modified nano-cellulose has a greater maximum adsorption capacity for Pb^{2+} (115 mg g^{-1}) than maleic acid-modified macro (20 mg g^{-1}), according to Vadakkekara et al. (2019). Furthermore, Hamad et al. (2020) prepared a hybrid nanofibers composite adsorbent membrane of modified cellulose nanofibers with modified hydroxyapatite for the removal of both lead (Pb) and ferrous (Fe) ions from simulated wastewater; the results demonstrated that this composite offers a higher adsorption capacity than either material alone. To achieve removal efficiencies of 99.7 and 95.47% for Pb (II) and Fe (III), respectively, the optimal conditions for the adsorption efficiency of Pb (II) and Fe (III) ions in wastewater were at the equilibrium time of 35 and 40 min, respectively, at $\text{pH} = 6$, room temperature, 0.1 gm of adsorbent, $V = 50 \text{ ml}$. Moreover, in a comparison study Sirviö and Ivanka (2020) compared the adsorption capacity of lignin-rich wood nanofibers and cellulose nanofibers produced from bleached cellulose fibers for lead and copper adsorption. Nanofibers of bleached cellulose pulp showed a lower adsorption capacity of copper and lead compared to lignin-rich nanofibers.



PTES ADSORPTION BY PAPER

The globe generates more than 300 million tons of paper annually, and demand is expected to double before 2030. Papers and cardboards account for 40% of municipal solid waste (Putro et al., 2019). Paper waste for PTEs adsorption has gained significant attention recently due to its low cost, high availability, and eco-friendliness. Several types of paper waste have been investigated for their adsorption properties, including recycled paper, newsprint, cardboard, and office paper. The properties of the paper waste, such as its surface area, pore size distribution, and chemical composition, significantly impact its adsorption capacity.

Recent studies have demonstrated the potential of paper waste as an adsorbent for PTEs. For instance, waste printing paper exhibited a high adsorption efficiency of 90% and metal uptake of less than 25 mg g^{-1} for evaluated metal ions (Moyib et al., 2017). Extraction of micro-fibrillated cellulose from wastepaper using simple sulfonation resulted in a 250% increase in adsorption capacity for lead (Pb) ions compared to pristine micro-fibrillated cellulose (Sridhar and Park, 2020).

In a study conducted by Chakravarty et al. (2008), newspaper pulp was used as an adsorbent for removing copper from effluent. The study found that the newspaper pulp adsorbent removed copper effectively, with a maximal loading capacity of 30 mg g^{-1} at an initial Cu concentration of 20 mg L^{-1} . The study also found that the adsorption of Cu onto the newspaper is a physisorption spontaneous endothermic process. Not only but also modified newspaper pulp with citric acid was used as an adsorbent: Pitsari et al. (2013) investigated the modification by 0.5 M and 1 M citric acid to improve lead adsorption, which increased adsorption capacity by 35% and 82%, respectively.

Additionally, paper sludge waste, a by-product of the paper industry, has been explored for PTEs adsorption. Adsorbents derived from de-inking paper sludge showed higher removal of Cu^{2+} than virgin pulp mill sludge (Méndez et al., 2009).

Several researchers have investigated the use of modified paper residue for the adsorption of potentially toxic elements. Using paper functionalized with polyethyleneimine (Setyono and Valiyaveetil, 2016) successfully removed nanoparticles, Ni^{2+} , Cd^{2+} , and Cu^{2+} cations, and Cr (VI) anions from polluted water samples. Coated polyethyleneimine paper demonstrated significantly higher adsorption capacities for the pollutants examined in this study compared to untreated paper; these capacities were observed for PTEs Ni^{2+} (208 mg g^{-1}), Cd^{2+} (370 mg g^{-1}), and Cu^{2+} (435 mg g^{-1}) ions compared to nanoparticles ($17\text{--}79 \text{ mg g}^{-1}$), and Cr (VI) (64 mg g^{-1}) anions.

Finally, the factors that affect the adsorption of PTEs onto paper waste have been extensively investigated, including the pH of the solution (Fawzy and Gomaa, 2020), the initial concentration of the pollutant, the contact time (Dehghani et al., 2016), and the temperature. Further research is needed to optimize paper waste's adsorption performance and explore its potential for large-scale applications.

PTES ADSORPTION BY WOOD MULCH AND SAWDUST

Mulch is a protective covering of material laid on top of the soil. Because of its low cost and simple availability, ground, shredded, or chipped wood is the most popular product in the mulch



industry. In addition, mulching provides soil erosion prevention, moisture conservation, and weed control (Soleimanifar et al., 2019). The quantitative examination of oak wood macromolecule content yielded data from many authors; according to Puech (1978), Kollmann and Fengel (1965), Herrera et al. (2014), the proportions of cellulose, hemicellulose, and lignin were 41%, 26.35%, and 25.71%, respectively.

Sawdust is a powdery by-product of woodworking processes such as sawing and milling. It can sorb PTEs in stormwater (Deng, 2020). Wood mulch and sawdust are effective adsorbents for various pollutants, including potentially toxic elements (PTEs). This review discusses some of the studies that have investigated the adsorption of PTEs by wood mulch and sawdust.

Different types of wood mulch have been investigated as adsorbents of PTEs from wastewater; in a study by (Jang et al., 2005), three types of mulch were used as sorbents to collect PTEs in urban runoff. The results revealed that hardwood bark mulch has the optimum physicochemical features for heavy metal ion adsorption. When the Hardwood mulch dosage was 4 g L^{-1} , and the pH was less than 6, more than 90% of the Pb (II) in the solution could be eliminated. Nevertheless, the required hardwood mulch dosage was more than 6 g L^{-1} , achieving better than 80% removal efficiency for Cu (II) and Zn (II). When the adsorption capabilities at pH 6.0 and 5.0 are compared, a slight variation indicates that the pH is unimportant.

In another study of hardwood adsorption of several contaminants by Ray et al. (2006), the results showed that chromium (Cr^{6+}), Copper (Cu^{2+}), cadmium (Cd^{2+}), zinc (Zn^{2+}), lead (Pb^{2+}), fluoranthene, naphthalene, butyl benzyl phthalate, 1,3-dichlorobenzene, and benzo(a) pyrene were all sorbed by hardwood mulch from a spiked stormwater pollutant sorbed mass depending on pollutant species, contact time, and initial concentration.

Furthermore, Iqbal et al. (2020) proved that mulches are an excellent source for removing PTEs from soil solutions, and the results showed that utilizing woodchips and compost in forest regions can create complexes with copper metal and transform it into a form that is not hazardous for crop plant growth.

Sawdust as an adsorbent of PTEs was studied by (Šćiban et al., 2007); the study focused on the efficiency of potential toxic elements removal by using sawdust with different quantities (1.25, 2.5, 3.75, 5, 10 and 20 g L^{-1}), the most significant removal efficiencies for Cu (II), Zn (II), and Cd (II) were 76.2, 37.5, and 31.9%, respectively.

Recently modifications on wood mulch and sawdust adsorbents were applied by different researchers; for example, the biosorption ability of natural and modified poplar, cherry, spruce, and hornbeam sawdust in removing PTEs from acidic model solutions was investigated by Kovacova et al. (2020): they studied the efficiency of alkaline modified sawdust for metal removal from model solutions at varying beginning concentrations of Cu (II) and Zn (II). Poplar treated by KOH had the maximum adsorption efficiency values for zinc (98.2% at pH 7.3) and copper (94.3% at pH 6.8). Sidhu et al. (2021) investigated other modifications, including using iron-based water treatment residuals coated wood mulch to reduce common contaminants in urban runoff. The results reveal that the unique adsorption media removed more Cu, Pb, Zn, and P than the typical uncoated mulch. Coated wood mulches were synthesized and evaluated in another study for removing PTEs and phosphorus (P) from synthetic urban stormwater (Soleimanifar et al., 2016). In batch experiments, the composite adsorption capacity was 97% lead (Pb), 76% zinc (Zn), 81% copper (Cu), and 97% phosphorus (P) for the tested synthetic stormwater (containing $\text{Pb} = 100 \text{ g L}^{-1}$, $\text{Zn} = 800 \text{ g L}^{-1}$, $\text{Cu} = 100 \text{ g L}^{-1}$, $\text{P} = 2.30 \text{ mg L}^{-1}$) at pH = 7.0.



Other reviewed modifications by Meez et al. (2021) of sawdust as an adsorbent material to enhance its selectivity and capacity involved various modifying agents, including (I) organic compounds (ethylene diamine, formaldehyde, epichlorohydrin, methanol, dyes); (ii) acid solutions (HCl, H₂SO₄, H₃PO₄, CH₃COOH, HNO₃, citric acid); (iii) mineral salts (NaCl, KCl, NaHCO₄); (iv) basic solutions (NaOH, Ca(OH)₂, KOH, Na₂CO₃); (v) phosphorylation treatment (CO(NH₂)₂ (urea) + H₃PO₄).

THE FATE OF USED CELLULOSE-BASED ADSORBENTS

Cellulose-based adsorbents are widely used in various applications such as water treatment, environmental remediation, and removing heavy metals and dyes from wastewater. However, the fate of these adsorbents after their use has become a significant concern due to the potential environmental impacts they may cause.

Incineration is a frequent way of disposing of used cellulose-based adsorbents. Unfortunately, this process can release harmful air pollutants like volatile PTEs (Xiong et al., 2019). Furthermore, the ash created by burning cellulose-based adsorbents may contain PTEs that are hazardous to human health and the environment. It causes secondary pollution through PTEs leaching into groundwater.

Another technique for getting rid of used cellulose-based adsorbents is landfilling. Unfortunately, the adsorbents can release hazardous chemicals into the soil and water nearby, endangering the environment. There may be issues with toxins seeping into groundwater since, in most situations, the contaminated cellulose-based adsorbent will contain high water content inappropriate for landfilling (Hubbe, 2022). In addition, the degradation of cellulose-based adsorbents in landfills can contribute to the creation of greenhouse gases, mainly methane, significantly contributing to climate change.

Scientists have investigated several techniques for reusing and recycling cellulose-based adsorbents to address these challenges, several researchers recycled the adsorbents using desorption techniques like heat or chemical treatment. This avenue of research was notably investigated by Liu et al. (2002), who tested the adsorption and desorption of Copper (II) using a spherical cellulose adsorbent. An aqueous solution of NaOH or HCl can be used to recover the Cu²⁺ ions that have been adsorbed on the adsorbent. The maximum recovery rate is nearly 100% using a 2.4 mol L⁻¹ HCl solution. Moreover, the adsorption capacity was lowered after 30 cycles of adsorption/desorption by only 7.2%. Therefore, the need for new adsorbents can be reduced, and waste can be reduced by reusing previously used adsorbents in various applications. For instance, the regeneration of adsorbents using the eutectic freeze crystallization process was explored by Hubbe et al. (2018).

Another strategy is to repurpose used cellulose-based adsorbents as value-added products. Adsorbents, for example, can be pyrolyzed to form biochar, which can be utilized as a soil amendment or a renewable energy source and for further adsorption processes. Agarwal et al. (2015) investigated the removal of azo dye using biochar from pyrolysis of cellulose-based materials municipal solid waste. Table 2 summarizes some PTEs cellulose-based adsorbents, and their treatment methods reported in the literature.

Finally, the fate of used cellulose-based adsorbents is a critical issue that requires careful consideration. Further research is essential to develop more efficient and sustainable methods for managing used adsorbents.



Table 2. Potentially toxic elements cellulose-based adsorbents and their treatment methods

Potentially toxic elements	Adsorbent	Treatment	Reference
Ni, Zn, Cd	Residues of pine sawdust, sunflower seed hulls, and corn residues mix	Immobilization of used adsorbents in clay ceramics	Simón et al. (2022)
Pb, Ni	Paper sludge waste	Sulfur treatment with K ₂ S solution	Wajima (2013)
Cu	Grape bagasse	Pyrolysis	da Silva et al. (2022)
Zn, Ni, Cu, Co, Cd	Succinic anhydride-modified mercerized nanocellulose	Regeneration	Hokkanen et al. (2013)
Cr	Pine sawdust and oak wood ash	Regeneration	Núñez-Delgado et al. (2015)
Cd, Zn, Pb, Cu	Paper and Mulch adsorbents waste	Incineration and Immobilization of adsorbent ash into mortar	Naser et al. (2023)

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