Wood-Composite for Wastewater Purification and Desalination

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The ecosystem has been seriously affected by sewage discharge and oil spill accidents. A series of issues (such as the continuous pollution of the ecological environment and the imminent exhaustion of freshwater resources) are becoming more and more unmanageable, resulting in a crisis of water quality and quantity. Therefore, studies on industrial wastewater purification and solar-driven seawater desalination based on wood composites have been widely considered as an important development direction. Generally, functional nanomaterials are loaded into the wood cell wall, from which lignin and hemicellulose are selectively removed. Alternatively, functional groups are modified on the basis of the molecular structure of the wood microchannels. Due to its three-dimensional (3D) pore structure and low thermal conductivity, wood is an ideal substrate material for industrial wastewater purification and solar-driven seawater desalination.

wood composites polymers wastewater purification seawater desalination

1. Introduction

Wood is composed of various tissue structures, cell morphologies, pore structures and chemical compositions. Therefore, it is a kind of polymer-based natural composite with a hierarchical and porous structure. Meanwhile, it also has obvious anisotropy, from the meter-level trunk to the decimeter- and centimeter-level wood fibers, millimeter-level annual rings and micron-level wood cells. Up to the cellulose nanofibrils, it has an extremely delicate and orderly multi-scale hierarchical structure [1][2].

Wood can be divided into coniferous wood and broadleaf wood. Coniferous wood mainly includes axial tracheids, wood rays, axial parenchyma and resin canals. Broadleaf wood mainly includes conduits, wood fibers, axial parenchyma and wood rays (a few types contain certain tracheids). It is these wood cells with different shapes, sizes and arrangements that form wood through an orderly and close combination. Then, they can create wood's unique pore structure [3]. According to their size, voids in wood can be divided into macropores, micropores and mesopores: (1) Macropores refer to pores that can be seen by the naked eye. Examples include wood cells (width: 50~1500 μm; length: 0.1~10 mm), vessels (20~400 μm), tracheids (15~40 μm) and intercellular canals (50~300 μm). (2) A micropore is a void with the order of magnitude of a molecular chain cross-section as the maximum starting point. For instance, the cross-section of the cellulose molecular chain is of the order of magnitude of a micropore. (3) Mesopores refer to voids with one, two or three dimensions in the nanometer scale (1~100 nm). For instance, there are marginal pores (10 nm~8 μm), simple pit pores (50~300 nm) and wood cell wall gaps (2~10 nm) in a dry or wet state and microfibril gaps (1~10 nm) in a swollen state in coniferous wood [4].

The wood cell wall is made up of approximately 45% cellulose (linear polymer composed of β-D-glucose) as the skeleton, approximately 30% hemicellulose (heterogeneous polymer composed of different types of monosaccharides) in a bonding role [5] and approximately 25% lignin (a complex, amorphous, 3D reticulated phenolic polymer composed of phenylpropane units) in a penetrating role. Wood is gradually assembled from monomolecular cellulose (~0.52 nm), elementary fibrils (2~3 nm), microfibrils (10~30 nm), macrofibrils (~10 μm) and cell wall lamellae (S1, S2 and S3 layers in primary wall and secondary wall) by inherent physical and chemical interactions [6]. After physical modification, chemical modification or physical/chemical combination modification, it will provide an important substrate and template for the bionic preparation of high-performance, high-value and multifunctional novel materials. The further development of functional and intelligent wood is bound to create an unlimited potential for novel material fields such as selective adsorption and separation, catalyst loading, water purification, seawater desalination, photoelectric devices and sensing devices [7][8][9].

2. Application of Wood Composites in Water Purification

2.1. Adsorption of Heavy Metal Ions

Because of their good solubility and stability, heavy metal ions in water exhibit the characteristics of high toxicity, non-degradation and biological enrichment in the ecosystem. If water containing heavy metal ions is discharged into the environment without treatment, it will cause serious harm to human health and the safety of other organisms [10]. Nowadays, the common methods to remove heavy metal ions from industrial wastewater include chemical precipitation, lime condensation, ion exchange, reverse osmosis and solvent extraction. However, they generally have problems such as complicated operations and high costs [11]. Therefore, an ideal choice is to treat heavy metal ions with adsorbents for the deep purification of water. Moreover, an adsorbent should meet the following standards: (1) low-cost and reusable; (2) effective and rapid; (3) selective and economically feasible [12]. The microstructure of wood contains a large number of hollow cells, which are interconnected and form interconnected channels, displaying a certain water flux. Moreover, wood is a typical multi-group ligand that can purify wastewater by adsorbing various heavy metal ions: (1) O– and COO– on the wood will react with heavy metal ions (Mn); (2) the negative polar bond in –OH, –NH, –OCH_3 and –C=O in the wood will generate electrostatic attraction with heavy metal ions; (3) –OH and –COOH in the wood will exchange ions with heavy metal ions, and H will be released into the water.

Sawdust is cheap and contains cellulose and lignin, which can absorb a variety of heavy metal ions. Therefore, it has a broad application prospect in the field of wastewater treatment. Ahmad et al. [13] ground sawdust into wood powder. Then, formaldehyde was used for the methylation reaction of wood powder to produce an adsorbent. The results show that the maximum removal rates for Cu^{2+} and Pb^{2+} are 99.39% and 94.61% when the adsorption material is in a solution with successive pH values of 7.0 and 6.6. Too high or too low a pH value will reduce the adsorption capacity of materials. This is because ion exchange and hydrogen bonding are the key to the efficiency of the removal of heavy metal ions by the adsorption material. In a water environment with a lower pH value, H competes with heavy metal cations for adsorption sites on adsorption materials. In a water environment with a higher pH value, OH^- will form soluble hydroxyl complexes with heavy metal cations, and the electrostatic interaction between heavy metal cations and adsorption materials will be weakened.

In order to improve the adsorption capacity, adsorption efficiency and selectivity of wood as adsorbent, other functional groups or inorganic nanomaterials can be further grafted or loaded in the wood channels. He et al. [14] prepared a 3D wood microfilter by modifying wood for the removal of heavy metal pollutants in wastewater. Specifically, a green deep eutectic solvent was used to remove lignin from beech wood. Then, carboxyl and sulfhydryl groups (-SH) were grafted on the surface of cellulose by sequentially using citric acid and l-cysteine. Finally, a 3D wood microfilter with an abundance of pores and adsorption sites was formed (**Figure 1**a). The adsorption kinetics and adsorption isotherms of heavy metal ions (Cu^{2+} and Cd^{2+}) on the 3D wood microfilter were systematically investigated. The results showed that the 3D wood microfilter had a fast adsorption rate and high saturation capacity for both Cu^{2+} and Cd^{2+}. Based on the advantages of easy multilayer assembly, a three-layer wood microfilter was designed to achieve the high flux rate (1.53×10^3 $L{\cdot}m^{-2}{\cdot}h^{-1}$) and high removal efficiency (>98%) for heavy metal ions in wastewater.

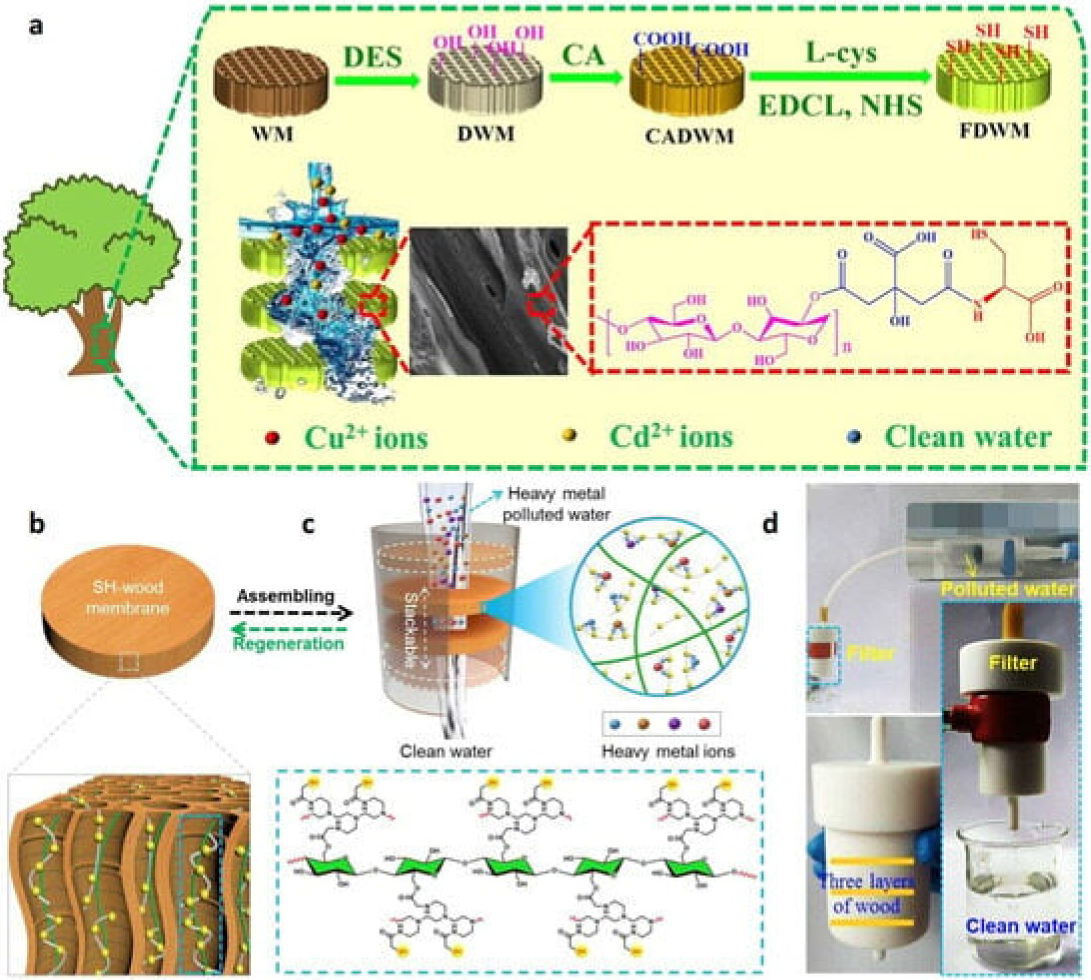

Figure 1. (**a**) A 3D wood microfilter for fast and efficient removal of heavy metal ions from wastewater. Schematic of SH-wood stacks for heavy metal ion removal from aqueous solution [14]. Reprinted with permission from Ref. [14]. Copyright 2020 ACS Publications Ltd. (**b**) SH-wood membrane, including a magnified drawing of its microstructure

and chemical composition [15]. (**c**) The multilayer device for large-scale heavy metal ion removal. The magnified schematic shows that the heavy metal ions can combine with −SH groups when the polluted water flows through the channels of modified wood [15]. (**d**) Photos of the experimental setup for the filtration of heavy-metal-polluted water and the clean water flowing out through the three-layer SH-wood membrane [15]. Reprinted with permission from Ref. [15]. Copyright 2023 ACS Publications Ltd.

Yang et al. [15] prepared a sulfhydryl functionalized wood (SH-wood) membrane with a three-dimensional mesoporous structure and low-tortuosity lumens. This SH-wood membrane serves as a multisite metal trap that achieves a high removal efficiency towards heavy metal ions from wastewater (**Figure 1**b,c). Benefiting from the unique microstructure of wood, the as-prepared membrane exhibits a high saturation absorption capacity of 169.5 $mg·g^{-1}$, 384.1 $mg·g^{-1}$, 593.9 $mg·g^{-1}$ and 710.0 $mg·g^{-1}$ for Cu^{2+}, Pb^{2+}, Cd^{2+}, and Hg^{2+}, respectively. Meanwhile, the SH-wood membrane can be easily regenerated at least eight times without apparent performance loss. Furthermore, an SH-wood filter with stacking multilayers was designed. Because of its high heavy metal ion absorption capability, the multilayer SH-wood filter can effectively remove diverse heavy metal ions from real wastewater, meeting the WHO standards and displaying a high flux rate of 1.3×10^3 $L·m^{-2}·h^{-1}$ (**Figure 1**d).

2.2. Disinfection and Sterilization

According to the statistics of the World Health Organization (WHO), approximately 1.6 million people die of diarrhea due to the lack of safe drinking water and basic sanitation facilities every year. Sterilization and disinfection of drinking water can effectively prevent diseases from being spread in water. The pore structure of wood has a natural barrier to larger colonies. After combination with antibacterial nanoparticles (NPs) (e.g., Ag NPs), a wood water filter with outstanding bacteria removal ability can be made. This is because Ag enters bacterial cells in the form of particles through endocytosis and is continuously released in the form of Ag. Specifically, Ag will cross-link or catalyze DNA molecules to form free radicals. Then, the proteins are denatured, the electron donors on DNA molecules are inhibited, and the DNA molecular chains break. In addition, Ag can combine with sulfhydryl and amino groups in cells, which will destroy the activity of cell synthetases. All the above procedures make bacteria and other microorganisms lose their ability to reproduce. When the bacteria die, Ag will be released and repeatedly perform the function of sterilization [16][17].

Electroporation sterilization technology applies a pulsed strong electric field to microorganisms and bacteria. It will destroy the cell membrane of bacteria and cause an osmotic imbalance inside and outside the cell membrane, eventually leading to the death of bacteria. There is no toxic by-product in the sterilization process. However, the high energy consumption and high risk of this technology limit its use in wastewater treatment [18]. Notably, researchers found that introducing 1D nanomaterials into conductive materials can solve the problems of energy consumption and safety [19]. Yang et al. [20] uniformly loaded Ag NPs into wood pores using the impregnation method, and the wood was further carbonized in a high-temperature tubular furnace. Then, Ag NP/carbonized wood membrane (3D Ag NP/WCM) composites with a three-dimensional mesoporous structure were obtained. The results show that the structure of nanofibers in carbonized wood is clearer. When a voltage is applied, the nanofibers will produce a peaking effect and greatly enhance the surrounding electric field, which can destroy the cell membrane of bacteria and lead to their inactivation. After electroporation, the damaged bacterial cells are more conducive to the invasion of Ag NPs in the carbonized wood and promote the sterilization process. The 3D Ag NP/WCM composite can be used in the condition of low voltage (4 V), low energy consumption (2 $J{\cdot}L^{-1}$) and high flux (3.8 × 10^3 $L{\cdot}h^{-1}{\cdot}m^{-2}$). It has a high bacteria removal rate (over 99.999%) and good stability (over 12 h). Compared with traditional electroporation sterilization technology, the wood composite not only avoids high energy consumption, but also reduces the safety risk of operation. It is a green, economical, fast, renewable and high-flux sterilization material for water treatment.

Nowadays, traditional methods for sterilization are chlorination disinfection, ozone disinfection, heavy metal ion disinfection, etc. They work by decomposing the organic matter, bacteria and microorganisms in water through a hydroxide reaction or peroxidation. However, they usually have some issues such as cancer-causing by-products, high cost and difficult maintenance. Compared with these traditional sterilization and disinfection methods, wood composites have the characteristics of high efficiency, simplicity, stability, low cost, environmental protection and so on, and they are basically not affected by the surrounding temperature and pH. In addition, wood is green and rich in cellulose, hemicellulose and lignin, and it does not produce secondary pollution or toxic carcinogens. Moreover, the wood microstructure can be resistant to many species (e.g., *Aspergillus Niger*, *E. coli*, *S. aureus* and *B. subtilis*) and achieve a water flux of up to 3.8 × 10^3 $L{\cdot}h^{-1}{\cdot}m^{-2}$. Wood composites are capable of removing at least 75% or even up to 100% of bacteria from wastewater. Therefore, wood composites have considerable development prospects in the field of sterilization and disinfection.

2.3. Removal of Aromatic Dyes

Printing and dyeing wastewater contains a lot of aromatic dyes, which are very difficult to remove. In addition, it also has the characteristics of dark color, high chemical oxygen demand (COD), high biological oxygen demand (BOD), complex and changeable composition, large discharge, wide distribution and difficult degradation. If the industrial wastewater is discharged without treatment, it will inevitably bring serious harm to the ecological environment due to its toxicity [21]. Therefore, removing these dye pollutants from water resources and wastewater is vital and important [22]. The natural pore structure of wood has a strong physical adsorption effect on aromatic dyes in wastewater. In addition, when the printing and dyeing wastewater flows through the pores of wood, its hydrodynamic effect is enhanced. In order to increase the time and opportunity of aromatic dye contact with active sites, functional nanomaterials or groups are loaded or grafted in the pore channels of wood.

Chen et al. [23] synthesized Pd NPs in situ in basswood microchannels by using a hydrothermal method to prepare a Pd NP/wood membrane. Specifically, cellulose, with rich hydroxyl groups, can immobilize Pd NPs; thus, the wood changed from yellow to black at first. This is because the plasma effect produced by Pd NPs fixed on the surface of a wood microchannel absorbs a lot of light. When wastewater containing MB flowed through the wood microchannels, MB was degraded by Pd NPs. The color changed from blue to colorless, and the MB degradation efficiency was over 99.8%. The interaction between MOFs and aromatic dyes can be used to treat different aromatic dyes in wastewater [24]. Guo et al. [25] used $ZrCl_4$, terephthalic acid and acetic acid as precursors for the in situ synthesis of UiO-66 MOF nanoparticles in three-dimensional mesoporous wood using the hydrothermal reaction method to obtain a UiO-66/wood membrane. Wood membrane filters for wastewater treatment can be obtained by changing the size and layers of this UiO-66/wood membrane according to actual needs. The results show that the flux of the filter assembled with three pieces of wood membrane is 1.0×10^3 $L \cdot m^{-2} \cdot h^{-1}$. The removal rates for cationic water-soluble aromatic dyes such as rhodamine 6G (Rh6G), propranolol and bisphenol A exceeded 96%, offering a rapid, multi-effect and recyclable method for removing aromatic dyes in this field. Wood has an abundance of nutrient transportation channels.

However, the efficiency of the removal of high-concentration aromatic dyes by conduits in wastewater is generally low, which is the same for wood-based devices with tracheid channels [26]. In solvothermal conditions, Cui et al. [27] introduced polyoxometalate-based metal–organic frameworks (POMOFs) into natural wood (POMOF/wood) for effectively removing aromatic dyes and capturing iodine. Keggin-type POM anions with a highly negative charge were encapsulated to adjust the charge of the UiO-66 MOF, and the charge overcompensation in the POMOFs allowed them to efficiently adsorb cationic dyes. Benefiting from wood's unique microstructure, the removal efficiencies of POMOF/wood towards MB and gentian violet (GV) (with a permeance of 1.0×10^4 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$) reach up to 94.07% and 95.23%, respectively.

2.4. Oil–Water Separation

Oil pollution in wastewater mainly comes from petroleum exploitation, the chemical industry, steel factories, coking workshops, gas generating stations and other industrial departments. Its mass concentration is generally 5000~10,000 mg/L. Most of these oils float on the surface of rivers and oceans and form oil films, resulting in a lack of oxygen in water bodies. Eventually, this results in the death of a large number of aquatic organisms. When oil washes up on a beach, it will cause serious harm to the waterfowls, shrimps, crabs and other creatures on the beach [28]. Wood also has a good effect on oily wastewater separation; its pit structure is very beneficial to the demulsification of oil–water emulsions. In the filtration and separation of oil–water mixtures, the wettability of the solid material plays an important role and has become an accelerator in this field. The key points in preparing special wetting materials are (1) the biomimetic construction of a hierarchical micro–nanostructure on the surface of a substrate with low/high surface energy or (2) directly using low/high-surface-energy materials to biomimetically construct a hierarchical micro–nanostructure on the surface of a substrate.

Superhydrophobic wood nanocomposites can be obtained by removing lignin from wood, further loading nanomaterials and then carrying out polymer backfilling and silanization treatment. Then, the expected goal for oil–water separation with high efficiency, high precision and high controllability can be achieved [23][26][29]. Zhao et al. [30] modified wood with polymethylsiloxane (POMS) to obtain superhydrophobic wood materials with a water contact angle (WCA) of 153°. POMS-modified wood has good oil absorption and oil–water separation performances. However, the accurate filtration efficiency and reusability of POMS-modified wood still need further discussion. Aside from cellulose, which is rich in hydroxyl groups, lignin and hemicellulose also exist in wood. They also contain $-NH_2$ and -OH groups, resulting in the good hydrophilicity of wood. When wood is soaked in water, a hydrophilic and oil-repellent water film is formed on its surface. When an oil–water mixture is dripped on a wood surface, the water permeates into the wood, while the oil is excluded, showing a good superoleophobicity underwater.

After selective removal of hemicellulose and lignin, the basic framework of cellulose with hierarchical high porosity and low density can be prepared easily [31][32]. Fu et al. [33] used a $NaClO_2$ solution to remove the lignin of balsa wood. After freeze-drying, a porous delignified wood template with high hydrophilic and oleophobic properties was obtained. Then, it was impregnated with epoxy resin/amine/acetone solution. After curing, a hydrophobic and oleophilic wood composite with a unique pore structure was prepared. The product shows an outstanding compression strength (263 MPa) and oil absorption effect (15 g/g), and it can absorb oil pollution on and under the surface of water at the same time. Wang et al. [34] coated one side of delignified wood with a dodecyl mercaptan solution. After ultraviolet radiation induction, a Janus wood membrane with asymmetric wetting properties and unidirectional water transmission was prepared, and it was suitable for selectively separating the mixtures of light oil/water and heavy oil/water (the separation efficiency was higher than 99.3%). Blanco et al. [35] directly used spruce (thickness: 1 mm) for oil–water separation. Under gravity, its flux (3500 $L \cdot m^{-2} \cdot h^{-1}$) and efficiency (>99%) for separating oil–water mixtures were investigated. After the loading of Ag NPs in wood, superhydrophilic and underwater superoleophobic wood nanocomposites were prepared, the surface hydrophilicity of which could be further improved. After in situ auxiliary modification of photothermal materials (graphene) and transparent hydrophobic materials on delignified and hemicellulose wood, Chao et al. [36] prepared a compressible and resilient photothermal wood aerogel.

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