

Review

Functionalized bio-based materials for water treatment: Advances in cellulose, lignin, melanin, and hybrid systems

Federico Paolino^a, Asanda Mtibe^b, Nomvuyo Nomadolo^b, Rosarita D'Orsi^a,
Jeannette J. Lucejko^a, Alessandra Operamolla^{a,*}, Maya Jacob John^{b,c}

^a Dipartimento di Chimica e Chimica Industriale, via Giuseppe Moruzzi 13, 56124 Pisa (PI), Italy

^b Centre for Nanostructures and Advanced Materials, CSIR, Pretoria, South Africa

^c Department of Chemistry, Nelson Mandela University, Port Elizabeth, South Africa

ARTICLE INFO

Keywords:

Bio-based materials
Water purification
Nanocellulose
Lignin
Melanin
Filtration
Electrospun membranes

ABSTRACT

Access to safe drinking water remains a global challenge due to increasing contamination by heavy metals, organic pollutants, dyes, pharmaceuticals, oils, and pathogenic microorganisms. In this context, sustainable and low-impact materials for water purification are gaining increasing attention. This review focuses on bio-based materials, specifically cellulose, lignin, and melanin, as well as their functionalized derivatives and hybrid systems, for water purification applications. These materials are integrated within a common framework, allowing direct comparison of their structure–property–performance relationships and highlighting potential complementary functionalities. Emphasis is placed on adsorption–filtration processes, membrane technologies, and electrospun fibrous systems.

Nanocellulose-based materials, including cellulose nanocrystals, nanofibers, and bacterial cellulose, are discussed in terms of extraction methods, surface functionalization, adsorption behavior, and performance toward a wide range of contaminants. Lignin-based systems, such as sorbents, hydrogels, foams, aerogels, and hybrid membranes, are examined with attention to the effects of chemical modification on adsorption capacity, selectivity, and reusability. Emerging melanin and melanin-like materials, particularly polydopamine-based systems, are also considered, highlighting their ability to bind metal ions and organic pollutants and their integration into multifunctional composites and membranes.

Across all material classes, the role of surface chemistry, porosity, and structural organization in determining adsorption performance is discussed, together with adsorption models, regeneration behavior, practical constraints, and the current level of technological maturity of the different material systems.

After an overview of biodegradability and sustainability aspects, the review critically examines key limitations, including long-term stability, regeneration efficiency, validation under realistic wastewater conditions, and scalability, which currently hinder large-scale implementation. These aspects are discussed to outline future research directions toward the development of effective and sustainable water purification technologies based on renewable bio-based materials.

1. Introduction

Globally, over two billion people lack access to safe drinking water, a crisis driven by industrial pollution, rapid population growth, diminishing freshwater resources, and climate change [1,2]. Contaminated water poses serious public health risks, contributing to the spread of waterborne diseases such as cholera, diarrhea, hepatitis A, and polio [3]. Inadequate water management further exacerbates exposure to unsafe

water sources. In response, the World Health Organization (WHO) has set ambitious targets to achieve universal access to safe drinking water by 2030 [3]. Meeting this objective requires the development of efficient, scalable, and sustainable water treatment technologies capable of removing a broad spectrum of contaminants, including heavy metals, organic pollutants, pathogenic microorganisms, pesticides, and polycyclic aromatic hydrocarbons [4].

To date, a wide range of advanced materials such as zeolites,

* Corresponding author.

E-mail addresses: alessandra.operamolla@unipi.it (A. Operamolla), MJohn@csir.co.za (M.J. John).

<https://doi.org/10.1016/j.ijbiomac.2026.153772>

Received 22 April 2026; Received in revised form 13 July 2026; Accepted 25 July 2026

Available online 28 July 2026

0141-8130/© 2026 Elsevier B.V. All rights reserved, including those for text and data mining, AI training, and similar technologies.

activated carbon, graphene-based materials, carbon nanotubes (CNTs), and metal or metal-oxide-based adsorbents have been extensively investigated for water purification applications [5–8]. While these materials often exhibit excellent adsorption efficiencies, their large-scale deployment is frequently constrained by high production costs, energy-intensive synthesis routes, limited recyclability, and concerns related to secondary pollution, such as nanoparticle leaching into treated water. These limitations have intensified the global search for alternative water treatment materials that are low-cost, environmentally benign, and derived from renewable resources, without compromising performance.

Among the most promising candidates are cellulosic materials and their derivatives, particularly nanocelluloses. These include cellulose nanocrystals (CNCs), cellulose nanofibers (CNFs), bacterial cellulose (BC), algae-derived cellulose particles, and tunicate cellulose nanocrystals (tCNCs) [9]. Nanocelluloses have demonstrated huge potential for the removal of various contaminants from water. This is due to their unique properties such as outstanding mechanical properties, large specific surface area, their availability and their amenability to surface functionalization. Furthermore, they exhibit biodegradability, eco-friendliness, and hydrophilic nature [4,9,10]. Functionalization with charged groups has further enhanced their capacity to adsorb various contaminants from water [9].

In parallel, lignin, the second most abundant biopolymer on Earth and a major byproduct of the pulp and paper industry, has gained increasing recognition as a value-added material for water purification [11]. Traditionally underutilized and largely incinerated for low-efficiency energy recovery, lignin represents an abundant and renewable source of aromatic carbon and functional groups. Although native lignin exhibits limited adsorption performance, targeted chemical modifications and nano structuring strategies have enabled the development of lignin-based adsorbents, membranes, and hybrid materials with significantly improved affinity toward metal ions, dyes, and organic pollutants [12]. The valorization of lignin for water treatment applications not only enhances purification performance but also aligns strongly with circular bioeconomy principles by converting industrial waste streams into high-value functional materials [13].

Beyond cellulose and lignin, melanin and melanin-like biopolymers have recently emerged as a novel and underexplored class of bio-based materials for water purification. Naturally occurring in animals, plants, and microorganisms, melanin exhibits exceptional metal-chelating ability, redox activity, and strong affinity for organic molecules [14]. Recent studies have demonstrated the effectiveness of fungal melanin nanoparticles, melanin–polymer composites, and water-soluble melanins derived from agricultural waste in removing heavy metals and organic dyes such as Cu(II) and malachite blue from aqueous systems [15]. These findings highlight melanin's potential as a multifunctional adsorbent and as a synergistic component in hybrid bio-based purification systems [16,17].

Conventional water treatment approaches including precipitation, coagulation/flocculation, ion exchange, photocatalysis, and membrane filtration have been widely implemented, yet each suffers from inherent drawbacks such as incomplete contaminant removal, high operational costs, limited reusability, and the generation of large volumes of secondary waste or sludge. Among available strategies, adsorption–filtration systems, which integrate adsorption processes with membrane or porous filtration, have emerged as particularly effective for the removal of toxic contaminants, including heavy metals, organic compounds, and microbial pollutants. Several comprehensive reviews have addressed individual classes of bio-based materials for water treatment, most notably cellulose- and lignin-derived systems, with emphasis on adsorption mechanisms, surface functionalization, or broader sustainability perspectives. Existing cellulose-focused reviews predominantly concentrate on nanocellulose synthesis routes, physicochemical properties, and adsorption performance toward specific contaminant classes, often treating membrane separation and adsorption as conceptually

distinct processes. Similarly, recent lignin-centered reviews have largely focused on lignin valorization, nanoparticle formation, or its role in composites and coatings, with comparatively limited integration of lignin-based materials into water purification architectures that combine adsorption and filtration functionalities. Reviews discussing melanin and melanin-like materials, on the other hand, remain scarce and are typically confined to biomedical, sensing, or niche environmental applications, with only fragmented coverage of their role in water remediation.

Despite the extensive literature on bio-based materials for water purification, the field remains fragmented, with most studies focusing on individual materials and model pollutants under simplified laboratory conditions. As a result, a clear understanding of how material properties influence performance under realistic conditions is still lacking. In addition, the relationship between material structure, functional properties, and purification performance is often discussed qualitatively but rarely examined in a unified framework. This review advances beyond existing literature by integrating cellulose, lignin, and melanin within a unified bio-based water purification framework, rather than treating these materials in isolation. In contrast to previous reviews that focus on single material classes, this work adopts a critical approach, highlighting limitations, and complementary functionalities across different bio-based systems. A particular emphasis is placed on the inclusion of melanin and melanin-like materials, which are still underrepresented in the context of water treatment but are here analyzed as design-relevant components alongside more established materials. Highlighting their complementary chemical functionalities, adsorption mechanisms, and structural roles, this review aims to address the represented gaps by providing a comparative analysis of cellulose-, lignin-, and melanin-based systems, with particular emphasis on structure–performance relationships and limitations relevant to real-world applications.

In addition, the review explicitly bridges adsorption studies with membrane-based and filtration technologies, providing a more application-oriented perspective that reflects the requirements of real-world systems. Rather than focusing solely on adsorption capacities, it evaluates how bio-based adsorbents are translated into scalable filtration architectures, including electrospun nanocellulose membranes, aerogels, foams, and layered hybrid filters.

This work incorporates a dedicated analysis of current limitations, including long-term stability, regeneration efficiency, fouling behavior, and scalability, supported by a comparative evaluation of representative systems. The review also proposes conceptual frameworks aimed at comparing the maturity, sustainability, and implementation readiness of different bio-based materials, providing a broader perspective on their transition from laboratory studies to real-world applications.

2. Adsorption mechanism

According to Norfarhana et al. [9], Reshmy et al. [4], Voisin et al. [18], and Norrahim et al. [19] adsorption process is the widely used and most reliable as well as convenient method to remove contaminants from the water. The adsorption relies on the binding of the analyte of interest (adsorbate) on the active sites of the adsorbent, as expressed in Eq. 1, and the process is modelled by adsorption isotherms and kinetic models (pseudo-first order and pseudo-second order) to assess the effectiveness of the adsorbent for the adsorption process [19].



Where A and B are concentrations of the adsorbate at equilibrium in the solution and adsorbent, respectively.

To quantify the adsorption process, Langmuir and Freundlich isotherms are often used as well as the combination of both isotherms known as Langmuir–Freundlich isotherms. Langmuir isotherm can be defined by Eq. 2.

$$q_e = \frac{Q_{\max} \cdot kA}{1 + kA} \quad (2)$$

Where q_e is the adsorbed amount of adsorbate at equilibrium, $Q_{\max}$ is the maximum adsorption capacity and k is the equilibrium constant.

The Freundlich isotherm is expressed by Eq. 3.

$$q_e = K_f A^{1/n} \quad (3)$$

Where K_f is the Freundlich constant related to adsorption capacity and n is the adsorption intensity.

The combination of Langmuir and Freundlich isotherms (expressed in eq. 4) is also used especially where there is a mixture of adsorbates in an environment such as wastewater and they are competing for selectivity in the active sites of adsorbent.

$$q_e = \frac{Q_{\max} \cdot (kA)^n}{(kA)^n + 1} \quad (4)$$

Maximum adsorption can be achieved by optimizing the adsorption parameters such as the pH of the solution, the concentration of the analytes in the solution, adsorption time, adsorption-desorption studies as well as adsorbent dosage [20]. Optimization of adsorption parameters also enhances the development of effective methods to remove contaminants from water.

The performance of bio-based materials in water purification is governed by three tightly interconnected factors: water transport, interfacial interactions, and structural organization. Actually, all these three aspects are intimately interconnected, as the wettability and permeability to water depend on the surface chemistry and on the morphology.

Water transport is primarily controlled by the porous architecture of the material, including pore size distribution, connectivity, and surface wettability. In highly porous systems such as nanocellulose-based aerogels and membranes, interconnected fibrillar networks promote capillary-driven flow and diffusion, enabling rapid water permeation. In contrast, in dense or poorly connected structures, mass transport becomes diffusion-limited, leading to reduced efficiency under dynamic conditions [21].

Interfacial interactions determine the actual removal of contaminants and depend strongly on both the chemical functionality of the material and the nature of the pollutant. In cellulose-based materials, adsorption is typically dominated by hydrogen bonding and electrostatic interactions, often enhanced by surface functionalization (e.g., carboxyl groups) [22,23]. In lignin-based systems, phenolic and aromatic moieties enable coordination with metal ions as well as hydrophobic interactions with organic compounds [24,25]. Melanin and polydopamine-based materials further expand this behavior through catechol and aromatic domains, which can promote strong binding via coordination, hydrogen bonding, and π - π interactions with aromatic pollutants [26].

These transport and interfacial phenomena are intrinsically linked to the structural design of the material. Hierarchical porosity, nanofibrillar organization, and the incorporation of multiple functional components in hybrid systems can significantly enhance accessibility to active sites while maintaining efficient mass transport. As a result, performance improvements are often not solely due to chemical affinity, but emerge from the synergistic coupling between structure, transport, and interfacial interactions.

3. Cellulose

Cellulose is the most abundant polymeric raw material in the world with an estimated annual production in the biosphere of 100 billion tons [27]. 6% of this production is presently utilized by industrial sectors such as packaging, cement, paper, textiles, cosmetics and automobiles.

Cellulose is the product of biosynthesis from plants, animals, or bacteria, and it is defined as a semi-crystalline polymer that comprises repeating units of D-glucopyranose ($C_6H_{11}O_5$) linked by the β -(1,4) glycosidic bonds. The biopolymer exhibits several characteristics, including good biocompatibility, minimal density, large physical properties, high strength, biodegradability, high crystallization and amenability to chemical modification[28]. Plants are the major producers, with plant cell walls comprising about 35–50% cellulose. The repeating units contain three hydroxyl groups that interact to form hydrogen bonds, which is particularly essential for the physical properties of cellulose as they govern the crystalline packing of the polymer. Different approaches, such as acid hydrolysis, mechanical treatment, and enzymatic hydrolysis, are used to extract nanocellulose from cellulose sources. These approaches produce particles with tailored properties such as crystallinities, surface chemistry, thermal expansion, and mechanical properties to suit specific applications. Nanocelluloses are fibers or crystals forms of cellulose with at least one dimension within the nanometer scale, and they retain most of the characteristics of cellulose. Nanocellulose is further classified into three categories: cellulose nanocrystals (CNCs), nanofibrillated cellulose (NFC), and bacterial nanocellulose (BNC).

Over the past several years, there has been a significant increase in the manufacturing of cellulose nanomaterials (CNMs) worldwide. In 2022, the market for nanocellulose was more than \$430 million, and it is expected to grow at a compound yearly growth rate of 24.7% through 2028 [29]. Additionally, compared to the favored carbon nanotubes (CNTs), promising for the automotive sector to reduce vehicle weight by as much as 60% while enhancing crash safety, the CNMs cost only US\$ 0.025/g and represent a more economical and sustainable modern-day material [30]. A market projection report by Cowie et al. [31] suggested that 40% of the body and interior components used in the annual manufacturing of 84.1 million cars could be made using CNMs, reducing the use of non-renewable products, lowering greenhouse gas emissions and the risks associated with managing solid waste because thanks to their biodegradability.

3.1. Structural and chemical features of cellulose

3.1.1. Cellulose nanocrystals

Acid hydrolysis of cellulose is the primary process used to produce CNCs, with hydrochloric and sulfuric acids being the most used acids to produce nanocrystals [32,33]. The amorphous area, primarily interacting with the reaction medium, is cleaved during hydrolysis, releasing the individual crystallites, or CNCs [34,35] as depicted by Fig. 1a. CNCs with a neutral surface are produced by hydrolyzing with hydrochloric acid and tend to aggregate in water [36]. Sulfuric acid hydrolysis is the most popular technique, since sulfuric acid yields more stable dispersions over a wide pH range ($pH > 1.0$); the so produced CNCs are characterized by the presence of sulfate groups, randomly formed from esterification of the primary C6-OH functionality of cellulose with sulfuric acid, that impart a negative surface charge to the crystals, allowing for better dispersion in water [37]. The crystallites generated are characterized as hard elongated tubulars, with length, width, and aspect ratios ranging between 50 and 500 nm, 2–5 nm, and 5–30 respectively, and 54–88% crystallinity index. The sources of cellulosic material and the hydrolysis reaction conditions determine the actual dimensions of the CNCs. Besides the acid hydrolysis method of producing CNCs, ionic liquid-mediated method, deep eutectic solvent method, enzymatic hydrolysis, TEMPO-mediated oxidation and oxidation-degradation methods have also been explored to produce CNCs [27].

In particular, enzymatic hydrolysis using endoglucanases has received great attention during the last decade, thanks to the significant yield improvements achieved both on cotton-derived cellulose pulp [36] and on cellulose-enriched biomass [38]. The problem of scarce dispersibility of neutral cellulose nanocrystals can then be solved by surface TEMPO-oxidation [39], that produces carboxylated CNCs and enable

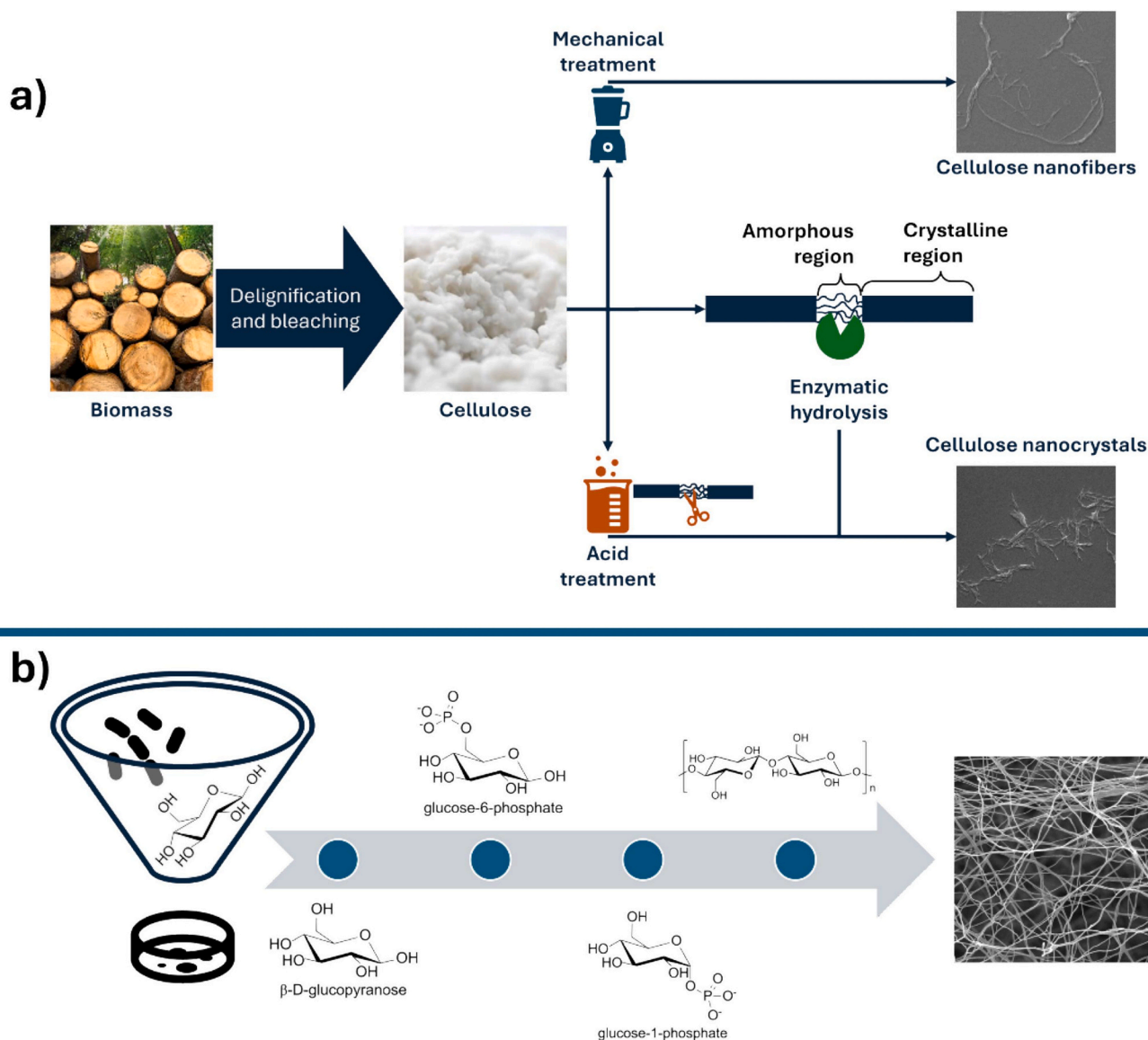


Fig. 1. Pathways for synthesizing cellulose nanomaterials. This Figure provides an overview of the pathways described in the review to prepare nanocellulose materials: a) starting from plants (wood) a chemical delignification step, followed by a bleaching treatment allows to isolate cellulose pulp, which can be then re-elaborated into cellulose nanofibers (CNFs) by mechanical defibrillation or into cellulose nanocrystals by enzymatic or acid hydrolysis; b) Bacterial nanocellulose (BNC) can be prepared feeding *Acetobacter xylinus* colonies with fermented sugars.

further functionalization [40,41]. Furthermore, neutral CNCs possess higher thermal stability than sulfated ones; this enables reactivity in thermally activated reactions in solvent free conditions which can potentially bring to energy savings of the functionalization process [42]. CNCs exhibit high surface areas reported to be ranging between 250 and 500 m²/g decorated with plenty of surface hydroxyl moieties. They also have a high tensile strength of 7500 MPa and a Young's modulus ranging from 100 to 140 GPa. These features of CNCs make them ideal reinforcing nanofillers for polymer nanocomposites [43].

3.1.2. Cellulose nanofibers

Interestingly, CNFs possess similar chemical properties to CNCs, but their physical properties are different due to the difference in extraction methods of the two, in the overall crystallinity and in the aspect ratio [4]. CNFs are predominantly extracted through mechanical defibrillation processes from cellulose source during which high shear stress exerted upon cellulose breaks down cellulose fibers into a slurry of fibril-like network of nanosized particles. Various mechanical methods are employed such as high-pressure homogenizing, ball milling grinding,

micro fluidization, cryocrushing, steam explosion and sonication techniques [44]. Typically, the type of mechanical extraction used determines the resulting particle aspect ratio, width, degree of branching, surface chemistry and charge. The nano particles are known to have 20–100 nm in diameter and micrometers lengths, lower crystallinity with respect to CNCs, due to the co-presence of amorphous regions along with crystallites. In addition, other impressive characteristics of CNFs include high elastic modulus, which is analogous to that of aramid fibers famously known for their use in ballistic protection armours, and thermal expansion on par with the glass typically used in automobiles and many other electrical applications [45].

Chemo-mechanical and enzymatic techniques are also well researched and frequently employed to separate cellulose nanofibers of refined weblike network with expanded textures with large surface area [46].

3.1.3. Bacterial nanocellulose

A growing body of research is focusing on BNCs because of the inherent unique spontaneous formation into nanostructured mesh

networks through fermentation of few genera of bacteria and for the high purity of the isolated material. These unique bacterial species are Gram-positive species like *Sarcina ventriculi* and Gram-negative species like *Acetobacter xylinum* (A. *xylinum*), *Rhizobium*, *Agrobacterium*, *Pseudomonas*, *Salmonella*, and *Alcaligenes*. A. *xylinum*, which has been renamed *Komagataeibacter xylinus* (or K. *xylinus*), is a strain of acetic acid bacteria characterized as rod-shaped aerobic bacterium with a 2 μm length and 0.6 μm width that produces bacterial cellulose with the highest efficiency with glucose as a feedstock, among all bacteria [47].

Four essential enzymatic steps are involved in the meticulous, and tightly controlled synthesis of BNC from glucose and the process is shown by Fig. 1b and it is as follows [48,49]. The process starts with an enzyme catalyzed reaction of a glucose molecule with the energy-carrying molecule called adenosine triphosphate (ATP), to form glucose-6-phosphate (G6P), this reaction activates the sugar molecule by turning it into sugar nucleotides, 2) This is followed by isomerization G6P to Glc-1-P (G1P) via phosphoglucomutase, thereby activating as anomeric diphosphate to enable glycosidation, 3) the activated G1P then polymerizes into β -1,4 glucan chains via the cellulose synthetase complex, leading to assemblage of repeating sugar units, and 4) BNC excretion from cytoplasm of cell to outer environment through cell membrane. K. *xylinus* creates a row of pores along the longitudinal axis of the cell and secretes cellulose in the form of microfibrils. The microfibrils emerging from the pores interweave to produce ribbons of cellulose which cover the medium's surface in a cellulosic mat. Each cell that is actively generating cellulose creates ribbons with sizes ranging from 20 to 100 nm [48]. Although the size of the fibers is mostly regulated by process variables such as temperature, stirring rate, and bacterium type.

Nanocellulose derived from plant cellulose (PC) and BNC share the same chemical structure; nonetheless, notable distinctions have been observed between them concerning physical attributes, purity, and macromolecular features. BNC is purer than PC because it contains no lignin, hemicellulose, or pectin, which permits a greater degree of polymerization and crystallization. Consequently, there's a growing consensus of replacing plant PC with BNC as lengthy and high costs of purification operations are avoided by substituting BNC for PC. However, the major point of contention is the costly capital investments and production. Additionally, the lack of well-organized scale-up processes resulting in low BNC yield is another constraint. This has resulted in the modification of technologies that increase the production of BNCs, such as cell-free systems, enhanced bioreactor-based bioprocesses, static intermittent fed batch strategies, and novel cost-effective substrates that might surmount the barriers associated with BNC production at industrial level.

Novel sugar rich substrates such as wastewater from several industries like sugarcane bagasse-processing industry, and agro-industrial wastes like pineapple waste and coconut water instead of conventional sugar sources is gaining significance as these offer the opportunity to explore underutilized biomass resources [50,51]. Therefore, cost-effective synthesis of BNC using innovative substrates, as well as upscaling production through better bioprocess technologies, necessitates further targeted study in order to completely commercialize BNC for industrial applications, particularly bio-based ones.

BNC has exceptional capacity to retain large volumes of water, unlike PC, due to high elasticity, conformability as well as water resistance. Additionally, the BNC fibers are finer compared to PC with estimated diameter range of 20-100 nm, rendering it an ideal filtration membrane.

3.2. Nanocellulose-based materials for water purification

3.2.1. Metals

Heavy metals are among the analytes of interest that make water not suitable for consumption. Although the surface of nanocellulose comprises alcoholic (OH) functional groups, they are not sufficient to stably coordinate heavy metals. Many studies, therefore, rely on surface

functionalization with negatively charged groups. It is also worth noting that determining the pH of the contaminated water is crucial in ensuring the overall efficiency of the remediation of heavy metals as it affects the surface charge of the adsorbent. Sulfuric acid hydrolysis is a very useful route to access negatively charged cellulose nanocrystals [37]. For instance, Baraskar et al. [52] obtained sulfated nanocellulose from rice husk and investigated the effect of the pH (ranging from 2 to 8) on the adsorption of various heavy metal ions, including Zn (II), Cd (II), As (III), Pb (II), and Sn (II). The results suggested that adsorption by CNC is strongly pH-dependent, with optimal adsorption capacity achieved in the range of pH 5–6 and with >50 min equilibration time. In addition, desorption and re-adsorption studies revealed that the adsorbent could be recycled for up to eight cycles. However, to achieve the optimal removal of heavy metals, the surface of the adsorbents should be more densely functionalized to have a stronger affinity for charged moieties thereby increasing adsorption capacity [53]. Indeed, surface sulfation typically achieves low substitution degrees. This is necessary to avoid cellulose degradation during the hydrolysis reaction [37].

Cationic cellulose nanofibers could remove Cr (VI) anions from water [54]. In this study, Kim et al. fabricated beads from carboxymethyl cellulose nanofibers (CMC-NF) grafted with polyethyleneimine with the aim of removing Cr (VI) from water. CMC-NF displayed an entangled morphology of fine nanocellulose as a consequence of the nanofibrillation induced by the carboxymethyl groups. The polymer showed the attitude to cross-link in the presence of cations forming beads; this could be taken as a proof of an easy manipulation of the material in water environment, where centrifugation or filtration would not be necessary to recover clean water after remediation. To induce cationization, the CMC-NF were mixed to a PEI (polyethylene imine) coating followed by glutaraldehyde (GA) crosslinking. The removal efficiency was evaluated by varying concentrations, time, and pH of Cr (VI) from 200 to 2000 mg/L, 0-360 min, and 1-10, respectively. The results showed that the PEI grafted CMC-NF adsorption capacity toward Cr (VI) was 1500 mg g⁻¹ under acidic conditions (pH between 1 and 2). The adsorption capacity toward Cr (VI) species was systematically >80%, against only 15.4% adsorbed by simpler CMC-NF beads, demonstrating the importance of the net surface charge of the adsorbent. In addition, the authors evaluated whether the results would fit the adsorption isotherm and kinetic models. The results suggested that the removal behavior of grafted CMC-NF followed Langmuir isotherm and pseudo-second order which indicates homogeneous adsorption and chemisorption, respectively. In the case of removal from drinking water, the removal efficiency was investigated in low concentrations of Cr (VI) (5 to 25 mg/L) revealing removal ability from 5 to 15 mg/L.

Sharma et al. [55] investigated the adsorption of Cd (II) from water using NOCNF, cellulose nanofibers from jute functionalized with carboxyl groups by a newly developed nitro-oxidation and extraction method, involving the mixtures of nitric acid and sodium nitrite as reagents. The extracted NOCNF presented high surface charge (ζ -potential -68 mV) and high carboxylate content (0.86 mmol/g), as determined by conductimetric titration. A static water contact angle of 38° revealed the high hydrophilicity of the sample, compatible with efficient water transport properties. The remediation mechanism of cadmium(II) nitrate was determined by characterizing the floc formed by mixing cadmium(II) solutions at different concentration with a 0.2 wt% NOCNF suspension and at pH 7.0. The authors inferred cross-linking occurring among Cd(II) addition, since cadmium can easily coordinate two carboxyl groups and the FTIR peak of the carboxyl anion, observed at 1581 cm⁻¹ for NOCNF, shifted to 1630 cm⁻¹ in the floc. The cross-linking induced gelification of the floc. The maximum adsorption capacity was 2550 mg g⁻¹, well above the theoretical quantity that would be adsorbed if only the carboxyl groups capture the cadmium ions. At low Cd²⁺ concentrations (< 500 ppm), the remediation process was dominated by the described coordination interactions with the carboxylate anions on the NOCNF surface, whilst at higher concentration, mineralization of cadmium hydroxide crystals occurred within the gelled NOCNF scaffold.

The combination of these two processes greatly enhanced the adsorption capacity of NOCNF.

Further results achieved in terms of heavy metals removal by nanocellulose-based systems are summarized in Table 1.

3.2.2. Oils and solvents

Oil and water are immiscible by nature, and as a result they create a thick sludge in wastewater treatment [14]. The use of adsorbents to separate oil and water is a well-known and established method. The key features that are required for the separation of oil and water are the porosity of the materials, and hydrophobic and/or oleophilic characteristics to entrap oil and separate it from water. In this context, aerogels possess intrinsic porosity and high specific surface area, and they can be used to separate oil from water, provided their surface is hydrophobic. Nanocelluloses can be used to prepare aerogels, which qualifies them to be absorbers of oil spillage in water environments, but they are intrinsically hydrophilic. Nevertheless, the surface of nanocelluloses can be tailored by chemical functionalization to impart hydrophobic properties. Shang et al. [70] created a superhydrophobic nanocellulose hydrogel crosslinking nanocellulose by an esterification reaction with 1,2,3,4-butanetetracarboxylic acid (BTCA) followed by freeze drying and surface modification with hexadecyltrimethoxysilane (HTMS). The findings indicate that the resultant aerogels, with a displayed porosity of 99%, were super-oleophilic and super-hydrophobic with a water contact angle of 151° . The maximum adsorption capacity of oil was 226 mg g^{-1} . More than 70% of the adsorption capacity was retained after 30 cycles of reusability and remained at 170 mg g^{-1} . Similarly, Zhang et al. [71] prepared microporous nanocellulose-based aerogels for the separation of oil and water. The microporous aerogels were prepared by blending nanocelluloses and chitosan followed by modification with quaternarized N-halamine siloxane monomer (QHS). The modification with these polymers resulted in the improved separation of different types of oil and water mixtures. In addition, the resultant microporous aerogels demonstrated antimicrobial properties against *S. aureus* and *E. coli* as they kill the bacteria in an oil/water environment and on the material surface. The resultant material can also be reusable without compromising the quality of the material.

Recently, a novel sponge composed of nanocellulose functionalized with stearyl chloride at 50° for 1 h was fabricated by freeze-drying to separate oil and water [72]. The sponge was superhydrophobic and displayed a water contact angle of 160° . Its oleophilic character made it very selective for oil removal. The adsorption of oil in water was very high (>25 times) in comparison to unmodified material. The resultant sponge material can be easily recovered by simple squeezing and reused at least 10 times without compromising its separation efficiency.

More research is required to explore and exploit nanocellulose-based materials for the separation of oil and water. These materials still present some challenges due to the difficulty of recovery of the used material for desorption and re-use. Some studies have evidenced the potentialities of magnetic nanoparticles incorporation into the absorbing nanocellulose to improve recyclability, but a definitive solution still needs to be found. Furthermore, freeze-drying, the process for preparing the aerogel /porous sponge, is an energy-intensive process, posing further economic and logistic issues.

3.2.3. Dyes

Organic dyes are widely used in the textile and paper industries as colorants. The process of colouring textiles, paper, and pulp requires a large amount of water [73]. After use, the contaminated water contains dyes that pose a serious threat to the environment and to human health. The comprehensive review published by Norfarhana et al. [9] reports that prolonged contact with a dye can cause diseases such as respiratory challenges and skin problems. Also, some dyes are mutagenic and carcinogenic. Removing dyes from water environments remains a challenge as most of them are water soluble, and they are non-biodegradable and highly resistant to chemicals and light [9]. The

Table 1

Adsorption of heavy metals using nanocellulose-based adsorbents.

Material	Heavy metal(s)	Results	References
Tannin-immobilized nanocellulose (TNCC)	Cu (II), Pb (II), and Cr (VI)	- Zeta potential revealed that TNCC had a negative charge and improved thermal stability. - The resultant TNCC displayed maximum adsorption of Cr (VI) as well as Cu (II) and Pb (II) at pH 2 and pH 6, respectively. - The adsorption capacities of Cu (II), Pb (II), and Cr (VI) were 51.85 mg g^{-1}, 53.37 mg g^{-1}, and 104.59 mg g^{-1}, respectively. - The adsorption fitted adsorption isotherms and follows pseudo-second-order kinetics.	[56]
CNC	Cu (II), Pb (II), and Zn (II)	- The removal efficiency of Cu (II), Pb (II), and Zn (II) by CNC were 88.75%, 81.18%, and 86.84%, respectively. - The adsorption followed pseudo-second-order kinetics. - The adsorption isotherms confirmed that the adsorption system was endothermic and spontaneous.	[57]
CNC-Bentonite nanocomposite	Pb (II) and Hg (II)	- The maximum adsorption of Pb (II) was higher than that of Hg (II). - The results fitted Langmuir isotherms and the system was endothermic	[58]
Dialdehyde nanocellulose (DANC)-carboxymethyl chitosan (CMC)-nanobentonite (NB)	Cr (VI), Co (III) and Cu (II)	- The maximum removal efficiency for Cr (VI), Co (III), and Cu (II) were 98.90%, 97.45% and 99.01%, respectively. - The results follow pseudo-second-order kinetics. - The adsorption system was endothermic, spontaneous, and feasible.	[59,60]

(continued on next page)

Table 1 (continued)

Material	Heavy metal(s)	Results	References
CNF-Fe ₃ O ₄	(Cr)(VI)	- Recyclability up to 6 cycles - The adsorption efficiency was 2.2 mg g ⁻¹	[61]
TEMPO CNF- ethylene diamine tetraacetic acid, aminopropyl-trimethoxy silane, 3-mercaptopropyl-trimethoxy silane, as well as graphene oxide (GO) and trimethylolpropane tris-(2-methyl-1-aziridine) propionate	Pb (II), Cr (III), Cu (II), Co (II), Ni (II), Mn (II), Zn (II), Sn (IV), Fe (III), Hg (II), Cd (II), and Zr (IV)	- The removal efficiency of about 87-100%. - Adsorption capacity of 208-718.5 mg g ⁻¹ . - Recyclability up to 5 cycles. - The results fitted Langmuir and pseudo-second-order kinetics. - The adsorption system was endothermic and spontaneous.	[62–65]
Carboxylated CNC-carbon dots (CDs)	Fe (III) and Pb (II)	- The removal efficiency for Fe (III) and Pb (II) were 69.4% and 98.2%, respectively. - The adsorption capacities for Fe (III) and Pb (II) were 98.3 mg g ⁻¹ and 442.0 mg g ⁻¹	[66]
CNC-grafted molecularly imprinted polymer	Pb (II) and Hg (II)	- The adsorption capacities for Pb (II) and Hg (II) were 27.55 mg g ⁻¹ and 161.31 mg g ⁻¹ , respectively. - The results followed pseudo-second-order kinetics and Langmuir isotherm.	[67]
Bacterial cellulose	V	- The adsorption capacity for V was 5.24 mg g ⁻¹ at a pH of 4.	[68]
CNC-TiO ₂	Pb (II)	- The adsorption capacity for Pb (II) was 0.552 mg g ⁻¹ . - The data indicated that Elovich kinetics and Baudu isotherms are suitable for Pb (II) adsorption.	[69]

chemical structure of some of the dyes discussed in this paragraph is presented in the Fig. 2. In recent years, there has been a paradigm shift toward the development of eco-friendly and sustainable adsorbents for the removal of dyes from water. Fabrication of nanocellulose-based adsorbents has become a research hotspot and their ability to adsorb dyes has been investigated. However, unmodified CNCs exhibit no adsorption of Congo Red [74], while unmodified CNFs were ineffective in reducing the concentration of Rhodamine B (RhB) in solution under the tested conditions [75]. The modification of nanocelluloses, achieved by blending with other polymers, reinforcing with nanoparticles, and functionalization to change the surface chemistry, is necessary to turn it into a dye adsorbent. The blend of nanocelluloses and chitosan has

shown potential for the removal of direct blue 78 dye [75]. The maximum removal efficiency was 91% when 14 g/L of concentration was used for 120 min under acidic conditions (pH 3.5). Under these conditions, chitosan is positively charged while the dye still shows negative charge due to its sulfonate groups. The results fitted Langmuir isotherms indicating that the adsorption process favors homogeneous adsorption as seen in other studies [76,77]. Thorat et al. [76] blended BC and PEI (polyethylene imine) for the removal of Congo Red dye and reactive red 120 dye. The maximum capacity was achieved under acidic conditions, and the removal capacity was 90% even after 4 recycling cycles. In addition, the adsorbent displayed antibacterial activities against *E. coli* and *S. aureus*.

Magnetic nanocellulose was also investigated for the removal of Rhodamine B (RhB) dye [75]. The magnetic nanocellulose was synthesized by in-situ preparation whereby Fe₃O₄ were prepared on top of CNF and activated with peroxymonosulphate (PMS). This active compound accelerates the degradation of dye. The results showed that when the active compound was absent there was no degradation of dye. However, when it was present about 95% of the dye was degraded over 300 min at room temperature.

Xue et al. [78] and Yap et al. [79] introduced the carboxyl group on nanocellulose to enhance its adsorption capacity toward methylene blue (MB), a positively charged dye. The maximum adsorption capacity of MB was around 925.93 mg g⁻¹ and removal efficiency was more than 93%. Other authors functionalized the surface of nanocellulose with trimethylammonium chloride [80] and 2-acrylamido-2-methylpropane sulphonic acid (AMPSA) [81] and studied the adsorption capacity of the dye. The adsorption capacity was 560 mg g⁻¹ for nanocellulose functionalized with trimethylammonium chloride and 357 mg g⁻¹ at 60 min, pH 6.0, and 30 °C for nanocellulose functionalized with AMPSA. The findings suggested that the adsorption process followed Langmuir isotherms and pseudo-second-order kinetics. Furthermore, the results indicated that the adsorbents can be reused for multiple adsorption-desorption cycles without compromising the adsorption capacity.

3.3. Nanocellulose based membranes for water treatment

Material architecture plays a key role in determining the practical implementation of bio-based sorbents. Fig. 3 provides a conceptual framework comparing conventional bulk sorbents with structured porous systems, highlighting how the introduction of internal channels and interconnected porosity enables continuous-flow treatment, facilitates regeneration, and improves compatibility with scalable water purification technologies. This distinction is particularly relevant when comparing laboratory-scale adsorption studies with technologies that can realistically be translated into continuous water treatment processes.

The commercially available membranes that are generally used as filters for water treatment are made from synthetic polymers. These include polyacrylonitrile (PAN), and polyvinylidene fluoride (PVDF) etc. [4]. These membranes have been extensively investigated and are economically viable, well-established, and efficient for the water filtration process [9]. This technology is most preferred for large-scale applications since it is easy to operate. To achieve optimal results, parameters and the properties of membranes should be optimized. The pollutants are excluded based on size. Membrane filtration is classified into conventional filtration, microfiltration, nanofiltration, ultrafiltration, and reverse osmosis depending on the membrane pore size and the pressure applied, as depicted in Fig. 4. Various contaminants such as yeast cells, bacteria, viruses, natural organic matter, sugars, and inorganics found are excluded by the size of the pores of the membrane. For instance, the pore sizes of the membranes used in microfiltration range between 100 nm and 10 µm and are suitable for filtering bacteria and viruses [4].

Even though these membranes have been used for decades, researchers have reported that they are susceptible to fouling due to their

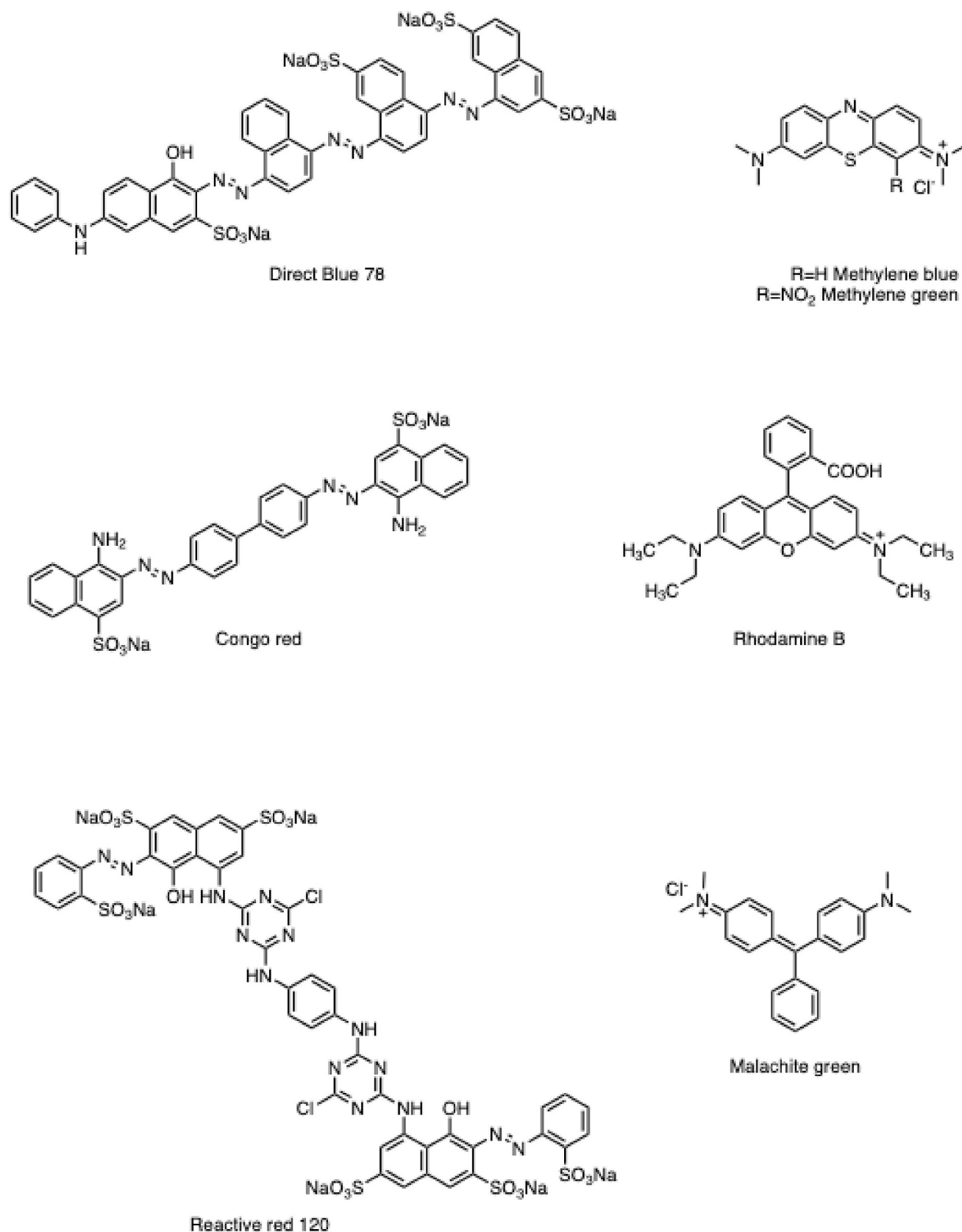


Fig. 2. Chemical structure of some water-soluble organic dyes. Azoic dyes, like Direct blue 78, Congo red and Reactive red 120, are not biodegradable in aerobic conditions and are negatively charged thanks to the presence of sulfonate groups. Other water soluble dyes present net positive or negative charge. Most of the adsorption mechanisms rely on the use of an adsorbent with overall opposite charge, especially when cellulose-based adsorbent are used. In the following paragraphs, strategies based on the use of lignin-based and melanin-based materials will be also presented. In those cases, π - π stacking interactions may be also exploited to remove dyes from contaminated water. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

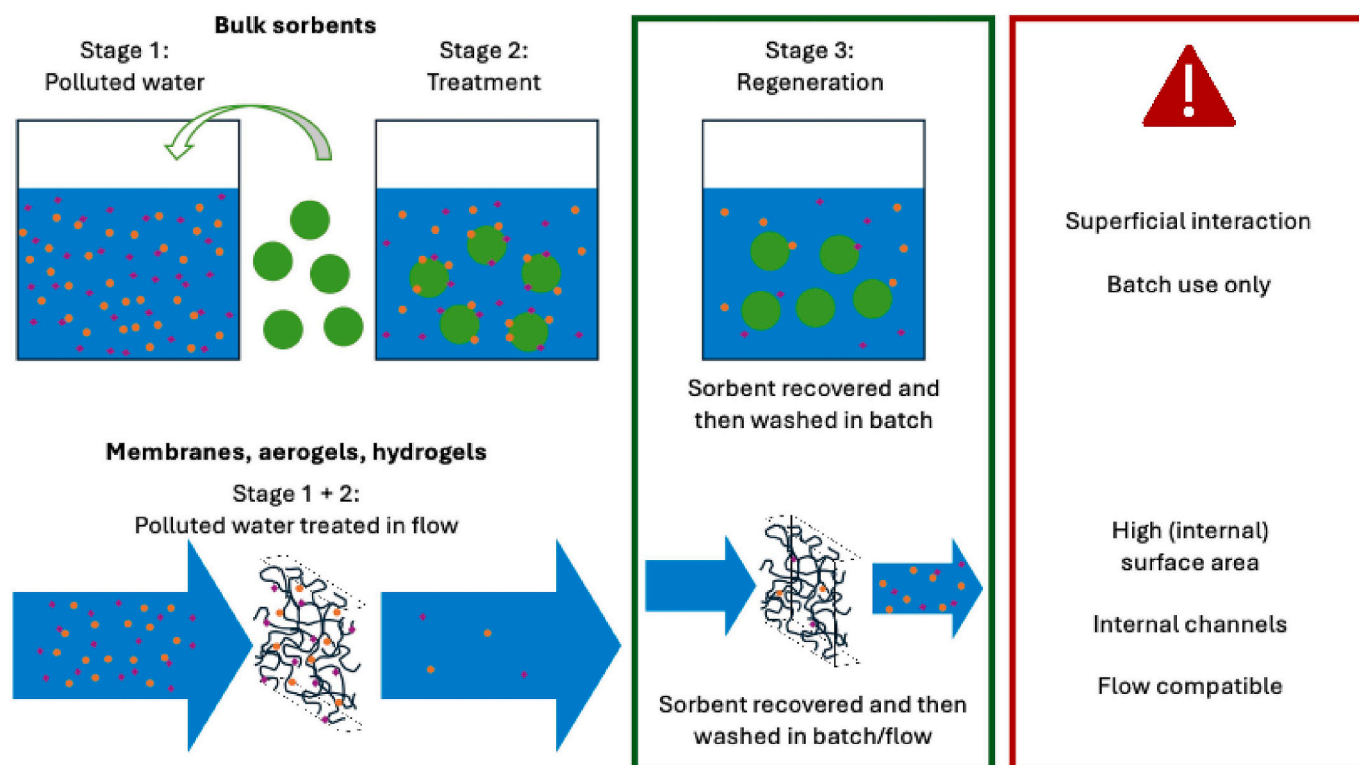


Fig. 3. Conceptual comparison between conventional bulk sorbents and structured bio-based water purification systems. Bulk sorbents are typically employed in batch adsorption processes that require subsequent recovery and regeneration steps. In contrast, membranes, aerogels, and hydrogels combine adsorption and water transport within an interconnected porous architecture, enabling continuous-flow operation and facilitating regeneration. Their high internal surface area and accessible pore structure improve sorbent utilization and may reduce material consumption, enhancing compatibility with scale-up and practical water-treatment applications.

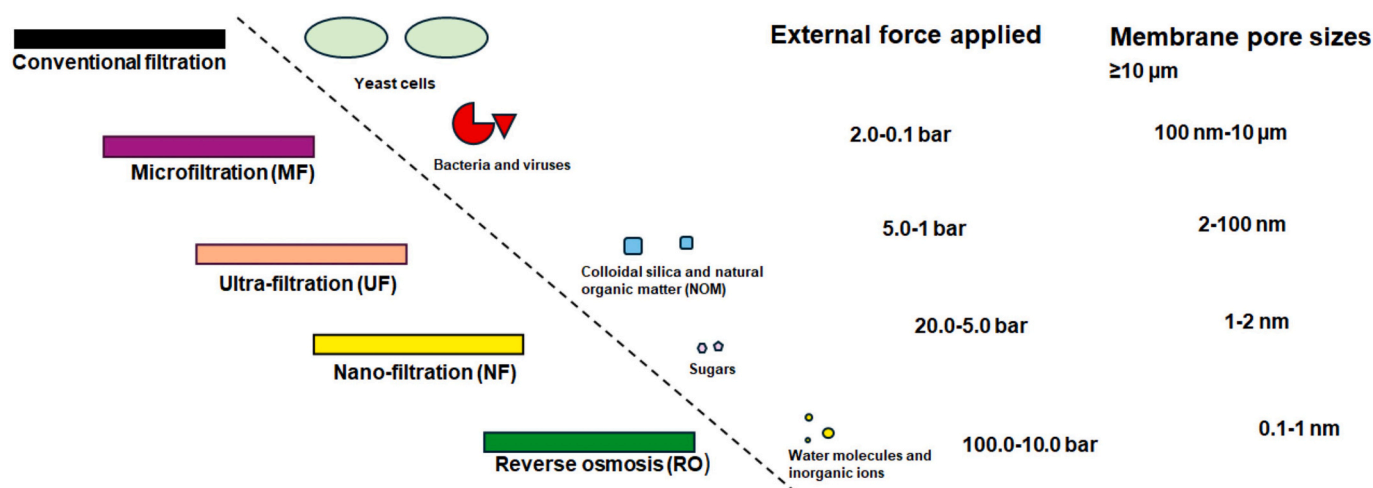


Fig. 4. Different types of membrane filters, type of contaminants removed, external force necessary for operating the filtration and average membrane pore size.

hydrophobic nature [82]. Due to the above-mentioned drawbacks, nanocelluloses are envisioned as a suitable alternative to synthetic polymeric membranes. This is due to their large surface area, porous hydrophilic nature, and tailorable surface [9]. Also, they have the potential to decrease fouling and improve permeability which are the major drawbacks of commercially available synthetic membranes. Nanocellulose-based membranes have been developed using various approaches, namely: i) vacuum filtration and coating, ii) solvent casting, iii) freeze drying, iv) impregnation of fiber mats with nanocelluloses, and v) interfacial polymerization [18]. The removal of contaminants from water mainly depends on the size exclusion rejection and

adsorption through active sites of the material [82]. For instance, Mautner et al. have prepared nanocellulose-based membranes by impregnating TEMPO-CNF into non-woven mats for the removal of heavy metals. This membrane displayed high adsorption of Cu (II) and high permeance which was attributed to the interaction between the active sites of TEMPO-CNF and Cu (II). The vacuum filtration method remains the suitable method used to develop nanocellulose membranes due to their simplicity and scalability [83]. The applications of nanocellulose-based membranes in contaminated water purification are shown in Table 2.

Unmodified CNF-based membranes possess higher mechanical

Table 2

Applications of nanocellulose-based membranes.

Materials	Contaminants	Processing method	Performance	Reference
Polydopamine/Bacterial Nanocellulose Hybrid	Rhodamine 6G (R6G), MB, and methyl orange (MO)	Solvent casting	Adsorption capacity of 8.7 mg g ⁻¹ at pH 4-7	[84]
CNC/Polyvinyl alcohol (PVA)-co-Polyethylene (PE) composite	Pb (II)	Solvent casting	Adsorption capacity of 471.55 mg g ⁻¹	[85]
CNF/Zn-Metal-organic frameworks (MOFs)	R6G and MB	Vacuum filtration	More than 85% degradation within 60 min, 5-cycle recycling	[86]
CNF-GO	Ag (I) and Cu (II)	Vacuum filtration	–	[87]
TEMPO-CNF/filter paper	Bovine serum albumin (BSA)	Vacuum filtration	Retention rate of 97.14% and flux of (46,279 $\frac{L}{m^2 \cdot h}$)	[88]
TEMPO-CNF/amyloid oligomers composites	Poly(ethylene glycol) (PEG), MB, BSA, Cr (III), (Fe (III), Ni (II), Cu (II)	Vacuum filtration	>90% rejection for molecules with >3 nm diameter. Metal ion removal efficiency from 45% and 89% for the composite	[89]
TiO ₂ /CNC	Pump oil	Vacuum filtration	Flux of 1887.4 ± 195.4 $\frac{L}{m^2 \cdot h \cdot bar}$ after UV-light irradiation for 4 h	[90]
AgNPs/CNC	Oil and water mixtures	Vacuum filtration	Flux >324 L m ⁻² h ⁻¹ bar ⁻¹ , oil rejection >99%	[91]
CNC/filter paper	Oil and water mixtures	Vacuum filtration	The separation efficiencies of >97%	[92]
Chitosan/CNC	Cu (II)	Solvent casting	Removal efficiency of >90%, adsorption capacity of >45 mg g ⁻¹	[93]

properties at low loadings than CNC-based membranes [34]. CNC-based membranes, on the other hand, can disperse easily due to their small sizes. These membranes are still facing the challenge of stability, high flux, and selectivity. However, the research on the development of nanocellulose-based membranes for water treatment is still in the infancy stage. New approaches to developing these materials such as in-situ functionalization of membranes and 3D printing should be exploited and explored to increase and optimize the use of the nanocellulose-based membranes for water treatment. The unique properties of nanocellulose-based membranes have displayed a high potential for commercial production at an industrial scale in the near future[94]. In the case of bacterial cellulose, although the research is promising, upscaling to an industrial scale is still a concern. Currently, there are no reports on the commercialization of BNC, CNF, and CNC-based membranes.

3.4. Electrospun mesh for water treatment

In this process, functionalized or unfunctionalized nanocelluloses are incorporated or coated onto the electrospun mesh to improve their effectiveness for water treatment. The research on the development of electrospun mesh for water treatment has been growing over the past decades due to their large surface area to volume ratio and porous structure [53]. Synthetic polymers such as polyvinylidene fluoride (PVDF), PVA, and biobased polymers (biopolymers) such as polycaprolactone (PCL), cellulose acetate, chitosan, poly(hydroxyalkanoates) (PHA) have been extensively investigated to form electrospun mesh for water treatment.

In a recent study, PVDF electrospun mesh reinforced with CNC and CNF has been developed to clean oily water and remove dye from wastewater [95,96]. The incorporation of low-loading CNC resulted in the improvement of the mechanical properties with an adsorption capacity of engine oil of 73.04 mg g⁻¹. In addition, the membranes were recycled 10 times without losing performance. In the case of PVDF membrane reinforced with CNF, the results showed that the adsorption capacity of crystal violet (CV) dye was 2.948 mg g⁻¹. Interestingly, the adsorption capacity improved to 3.984 mg g⁻¹ when CNF was functionalized with Meldrum's acid. A similar trend was observed in the adsorption of Fe (II) in the same study. The functionalization of CNF with tea leaf microparticles and incorporation into the PVA electrospun

membrane has also resulted in the improved adsorption capacity of Cr (VI) [97]. This was due to the presence of carboxylate, aromatic carboxylate, phenols, and oxy functional groups which facilitate the adsorption of heavy metals.

Biobased chitosan membrane reinforced with phosphorylated nanocellulose has shown a potential to remove Cd (II) water [98]. The adsorption capacity of 62.3 mg g⁻¹ was achieved, and the data fitted Langmuir and pseudo-second-order kinetics. Electrospun membranes obtained from synthetic biobased polymers such as PHA, and PCL reinforced with nanocellulose were also investigated for water treatment. The membranes fabricated from PCL and CNF displayed porous structures and were able to remove 75% to 99% of Fe (II) and Cr (IV) [99].

4. Lignin

Lignin is a highly complex, non-crystalline biopolymer and a key constituent of plant cell walls, where it provides structural stiffness and resistance to biological and chemical breakdown [100]. It consists of an irregular three-dimensional network of phenylpropanoid building blocks linked by a variety of ether and carbon-carbon bonds. Owing to its aromatic-rich and chemically intricate architecture, lignin represents an attractive feedstock for the production of functional carbon-based materials. It is the second most abundant biopolymer on Earth, after cellulose, and constitutes the largest renewable reservoir of naturally occurring aromatic structures. Lignin represents a waste of the pulping processes, such as wood processing and pulp and paper production. It is biosynthesized from the radical polymerization of C6C3 alcoholic monomers, named monolignols and reported in Fig. 4a, which occurs in plant cells by enzymatic dehydrogenation. Lignin has an amorphous structure, presenting different kinds of connections among monomers, mostly based on ether or carbon-carbon linkages, as described in Fig. 5.

Lignin possesses a variety of hydroxyl groups, that are important in determining its physicochemical characteristics such as solubility, hydrophilicity, reactivity, and functionality. The use of lignin has been limited by the scarce knowledge of its molecular structure and properties, but nowadays advanced techniques help researchers to gain more insights about this interesting biopolymer [101,102]. Some reports indicate that, among novel biosorbents derived from biomass resources, lignin-based materials possess good adsorption property to both toxic

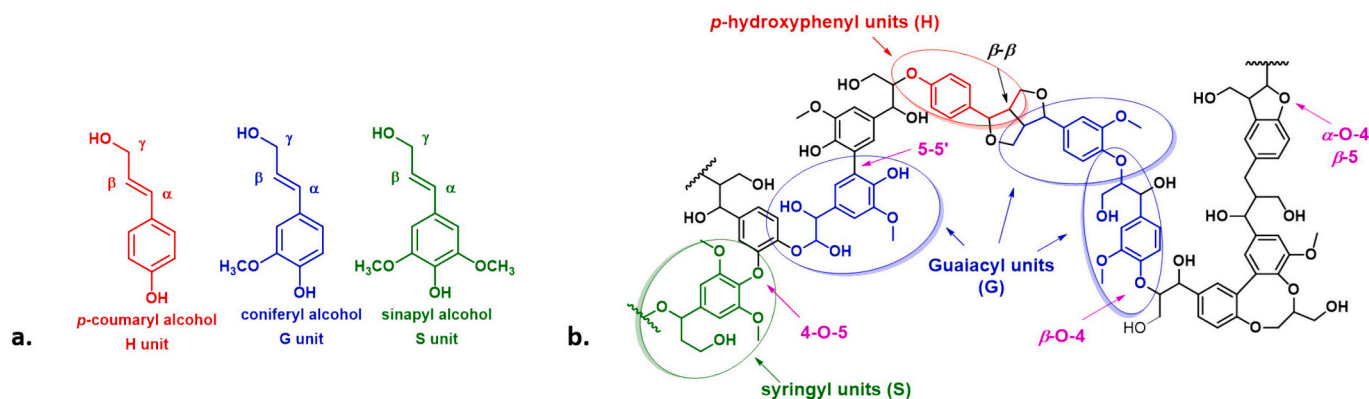


Fig. 5. Conceptual description of the chemical structure of lignin. a. C6C3 alcoholic monomers of lignin structure, namely *p*-hydroxyphenyl (H), guaiacyl (G) and syringyl (S) units; b. most common intermonomer fusions occurring in the lignin structure, such as ether linkages between oxygen in position 4 of one monomer and carbon 5 of the other monomer (4-O-5); oxygen in position 4 and the carbon β or α of the aliphatic chain of another isomer (β -O-4; α -O-4) or carbon-carbon linkage between carbon number 5 of one monomer with the 5 or with the β of the chain of another unit (5-5' or β -5).

dyes and heavy metals ions in polluted water. Adsorption mechanisms in bio-based materials are strongly influenced by interfacial interactions. For instance, lignin-based sorbents exhibit affinity toward metal ions through coordination bonding with phenolic and carboxyl groups, or can interact with aromatic pollutants via π - π stacking and hydrogen bonding, leading to enhanced adsorption performance. Therefore, lignin holds an enormous potential as a cost-effective contaminants remover from aqueous media. Several applications of this interesting material in advanced (nano)technology and contaminated water treatment and purification have been described, using lignin hydrogels, lignin supported metal/metal oxide nanocomposites, or functionalized lignin [103]. Furthermore, adsorption, catalytic reduction and (photo)degradation of a variety of pollutants have been demonstrated [104]. Here, we present an overview of the lignin-based materials and their adsorption capabilities and the recent progress in their utilization.

4.1. Lignin based materials for water purification

Although lignin in its native form can potentially act as a sorbent for water purification, its performance is generally limited, because the effective number of functionalities exposed to water is poor, unless the specific surface area is increased by chemical manipulation or fabrication methods. However, lignin is a useful carbon neutral starting material to prepare porous foams or hydrogels, which can be exploited in water filtration economy. Furthermore, lignin's extraction process can dramatically modify its chemical structure, and the content of functionalities which may be exploited for the adsorption of pollutants, such as carbonyls, phenols, carboxylic groups or other hexogenous groups introduced by the chemical treatment that may be charged such as sulphonates, etc. Finally, to enhance its adsorption capacity and selectivity, various chemical modifications can be introduced to the lignin molecular framework. The most straightforward approach involves exploiting the reactivity of phenolic hydroxyl groups, which represent the main functional moieties of lignin. More advanced strategies rely on crosslinking reactions to produce highly porous materials, such as hydrogels, capable of trapping pollutants within their three-dimensional network through a combination of physical entrapment and chemical interaction. These porous structures can be obtained either from lignin alone—using a crosslinker—or from blends of lignin with natural or synthetic polymers, depending on the desired mechanical, chemical, or adsorption properties [12].

Another approach involves the preparation of lignin micro- or nanoparticles via precipitation techniques. Such particles can be further functionalized to improve sorption efficiency, for example by incorporating magnetic cores that facilitate recovery and reuse [12,105].

Finally, lignin can also serve as a precursor for the production of

activated carbon materials. Although these exhibit good sorption performance, they are not included in the present review, as the preparation processes—such as applied pyrolysis—lead to extensive structural transformations that eliminate the characteristic functionalities of lignin.

4.1.1. Metals

The removal of heavy metals from water is one of the most investigated applications of lignin-based sorbents. The abundance of hydroxyl, methoxy, and carboxylic groups in lignin provides numerous coordination sites for metal ion binding. Different chemical modifications—such as oxidation, crosslinking, and hybridization with inorganic components—have been explored to enhance sorption capacity, selectivity, and recyclability.

A lignosulfonate-based hydrogel covered with a polypyrrole layer was prepared through a crosslinking-polymerization strategy with *N*-isopropylacrylamide and itaconic acid at 70 °C under argon atmosphere. The hybrid system exhibited excellent sunlight adsorption (96%) and high stability across a wide pH range (1–14). Hydrogels possess very good water transport properties, and the presence of polypyrrole as light absorber enables a new material for solar evaporators, devices which can effectively deliver fresh water upon evaporation and recondensation cycles. In solar evaporators, water evaporation enthalpy may decrease in the micro-network of hydrogel thanks to the interactions between hydroxy groups of lignosulfonate and water molecules, enabling efficient desalination and removal of Cu(II), Cr(VI), and Co(II). The hydrogel's performance was attributed to the uniform microporosity of the lignin matrix whilst the polypyrrole coating ensured efficient heat transfer and durability under harsh conditions [106].

A lignin-polyurethane foam was modified with polydopamine (LPUF/PDA/PA) and phytic acid to enhance hydrophilicity and oil-phobicity for use in underwater filtration devices. The foam displayed uniform porosity and high compressive strength. Originally designed for oil-water separation, the material also demonstrated high Cu (II) adsorption capacity (98.2 mg g⁻¹) after 200 min of contact, confirming its multifunctional potential for water purification [107].

An electrospun membrane composed of alternating layers of silk fibroin and a lignin-polyacrylonitrile (PAN) mixture was fabricated through sequential deposition to form a five-layer structure [108]. The membrane demonstrated pronounced hydrophobicity, allowing efficient water repellence and good adsorption of apolar pollutants. The material was further tested for heavy metal retention, showing rejection rates of 35.5% for Cu(II) and 84.6% for Cd(II), confirming its selectivity and efficiency in separating specific ions [108].

Lignin-based nanotraps (LBNT) were synthesized by emulsifying lignin, diethylenetriamine, and formaldehyde in liquid paraffin with

Span 80 as surfactant, obtaining decorated lignin nanoparticles. The nanoparticles were then esterified with CS₂. The xanthate nanoparticles showed strong pH-dependent adsorption of Ag(I), Pb(II), and Cu(II). Optimal adsorption was achieved between pH 5 and 7, with equilibrium reached within 30 min. The nanotraps exhibited >99% adsorption efficiency in multi-ion systems containing 5 ppm of Ag(I), Hg(II), Cd(II), Pb(II), Cu(II), and Zn(II), confirming their potential for simultaneous removal of multiple contaminants [109]. At the same time, particles loaded with silver displayed very interesting antibacterial properties versus *E. coli* and *S. aureus*.

A porous sorbent synthesized from lignosulfonate, citric acid, and acrylic acid using *N,N'*-methylenebisacrylamide as crosslinker exhibited maximum adsorption capacities of 250 mg g⁻¹ for Cu(II) and 300 mg g⁻¹ for Pb(II) at pH 7, reaching equilibrium after 60 min. The material retained about 80% of its initial performance after five adsorption-desorption cycles [110].

Kraft lignin, a technical lignin prepared through the Kraft pulping process, possesses abundant phenolic groups, carboxylic acids and can potentially be used to adsorb metals or other pollutants. A silica-Kraft lignin hybrid was prepared via the sol-gel method to obtain a composite with a three time capacity to adsorb pollutant with respect to the isolate starting materials. The silica-lignin hybrid demonstrated 63% Co(II) removal after 90 min of contact, with optimal adsorption occurring at pH ≥ 7 in 1.5 h and poor efficiency observed under acidic conditions (pH < 7), demonstrating a potential use in the recovery of cobalt from spent batteries. The system demonstrated a flexible porosity, depending on the hydration degree of the silica nanoparticles, that could break clusters in water and expose higher surfaces to contaminated solutions. The fast adsorption kinetic observed for Co (II) was justified accounting for an easier interaction of the material with metal ions, due to favorable dimension or hydrophilicity of the material, or to very favorable lignin phenolic OH- Co(II) coordination interactions. However, increasing the temperature led to a marked decrease in adsorption capacity and reusability tests conducted through desorption cycles with inorganic acids showed that the material experienced a significant performance loss after the second regeneration cycle. [111].

Dual-modified lignin obtained through sequential reactions with formaldehyde/CS₂ and chloroacetic acid exhibited high Pb(II) adsorption efficiency, with nearly 99% removal at pH 6 [112]. The process was endothermic, with higher temperatures enhancing adsorption efficiency. Equilibrium was reached after approximately 2 h. Competitive adsorption tests using Cu(II) and Mn(II) revealed a preference for Pb(II), although the presence of other metals slightly reduced its uptake. Reusability tests indicated a 30% decrease in capacity after three cycles.

A lignin-based sorbent functionalized with quaternary ammonium groups and decorated with Zn(II) was recently reported for As(V) removal [113]. The material was synthesized by reacting lignin with 3-chloro-2-hydroxypropyl trimethylammonium chloride, followed by precipitation with K₃Fe(CN)₆ and HCl, and subsequent treatment with ZnCl₂. The sorbent showed optimal performance at pH 5, achieving ~70% As(V) removal with a maximum adsorption capacity of 72.6 mg g⁻¹, and adsorption equilibrium was reached in about 45 min. Reusability tests demonstrated good stability, with only an 11% decrease in capacity after five adsorption-desorption cycles [113].

Magnetic, functionalized lignin-based nanoparticles have also been reported for the adsorption of heavy metals [114]. Fe₃O₄ particles with and external silica shell Fe₃O₄@SiO₂-NH₂ where coupled with carboxymethylated lignin in the presence of epichlorohydrin. For comparison, non-silica coated modified lignin particles and unmodified lignin particles were also prepared. Adsorption studies of Pb(II) and Cu(II) over the pH range 2-6 revealed maximum performance at pH 5, with capacities of 90 mg g⁻¹ for Pb(II) and 60 mg g⁻¹ for Cu(II). Notably, equilibrium was reached within 30 s. However, the particles exhibited instability in acidic conditions, preventing efficient cleaning and reuse [114].

Overall, these studies confirm that lignin is a versatile platform for

designing sorbent materials for heavy metal removal. By applying targeted chemical modifications, lignin-based systems can achieve high adsorption capacities, rapid kinetics, and in some cases multi-ion selectivity. Nonetheless, issues such as stability under acidic conditions and limited recyclability remain significant challenges, highlighting the need for further optimization before industrial-scale applications can be realized.

4.1.2. Oils and solvents

Lignin-based materials have also been studied for oil-water separation and the removal of organic solvents from contaminated water. Due to their tunable surface chemistry and compatibility with polymer matrices, lignin-derived foams, aerogels, and composites can combine hydrophobicity and oleophilicity, making them suitable for absorbing oils and non-polar contaminants while maintaining mechanical robustness and reusability.

The previously described LPUF/PDA/PA foam was also employed for oil-water separation [107]. Mixtures of *n*-hexane, cyclohexane, toluene, and pump oil with water were filtered through the foam. In all cases, removal of the organic phase exceeded 99%, and performance was maintained over 10 cycles, demonstrating the robustness of the system [107].

A lignin-polyurethane (LPU) foam for oil-water separation was fabricated by acetylating Kraft lignin with acetic anhydride, followed by mixing with polypropylene glycol (PPG), dibutyltin dilaurate, cyclopentane, and polymeric 4,4'-diphenylmethane diisocyanate. For comparison, non-acetylated Kraft lignin was also used. The presence of non-acetylated lignin led to significant alterations in porosity due to particle aggregation, whereas acetylated lignin maintained a uniform structure up to 20% loading. Adsorption tests with various oils showed the best performance for crude oil and olive oil: ~4 g g⁻¹ for the PU foam and ~8 g g⁻¹ for the PU foam with 20% acetylated lignin. Reusability studies further demonstrated that the acetylated lignin foam at 20% retained over 90% of its performance after 50 cycles [115].

An aerogel for oil-water separation was prepared by aminating lignin with diethylenetriamine (ratios 1:1, 1:2, 1:3), then blending it with poly(vinyl alcohol) (PVA). To enhance hydrophobicity, methyl trimethoxy silane was deposited by chemical vapor deposition (CVD). Since the nitrogen contents found in the composites were similar independently of the ratios between lignin and amine used during the preparation, the 1:1 formulation was selected for economic reasons. The aerogel was highly hydrophobic at any pH in the range 1-13. It displayed adsorption capacities ranging from 2 to 11 times its weight depending on the solvent. Removal of organic solvents from water exceeded 90%, and the aerogel maintained excellent performance over 10 reuse cycles [116].

Polypropylene(PP)-lignin sponges were fabricated by dissolving polypropylene in a decalin/1-butanol mixture and adding lignin at different ratios before cooling to form the sponge. Incorporation of lignin produced rougher pore surfaces compared to neat PP sponges. Increasing lignin content reduced the water contact angle but values remained >90°, confirming hydrophobicity. At 10% lignin, the sponges exhibited the highest retention for soybean, engine and lubricating oils (>90% for all). Reusability tests showed a decline in oil uptake during the first three cycles (from ~100% to ~80%), but performance stabilized in subsequent cycles [117].

Overall, the reported studies demonstrate that lignin-based materials are highly effective for removing oils and organic solvents from water, offering cleaner and more sustainable solutions for water purification.

4.1.3. Dyes

The aromatic and functionalized structure of lignin offers strong interactions with dye molecules via π - π stacking, hydrogen bonding, and electrostatic attraction. Numerous studies have demonstrated the effectiveness of lignin-based sorbents—often chemically modified or combined with other biopolymers—in removing both cationic and

anionic dyes under optimized pH conditions.

The previously presented lignin–polyurethane foam LPUF/PDA/PA was also employed for water soluble dye adsorption, specifically Methylene Blue (MB) and Rhodamine B (RhB) as model compounds (chemical structures shown in the Fig. 2). The material achieved adsorption capacities of 67 mg g^{-1} for MB and 96 mg g^{-1} for RhB, maintaining 85% of its original performance after ten adsorption–desorption cycles carried out with ethanol washing [107].

The same electrospun membrane composed of silk fibroin and lignin–PAN layers used for removing cadmium and copper, demonstrated remarkable efficiency in removing cationic dyes, including Crystal Violet, Methylene Blue, and Brilliant Green. Removal exceeded 99% for all three dyes, confirming the membrane's high affinity toward positively charged molecules [108].

An amine-crosslinked lignin sorbent was synthesized by reacting kraft lignin with 2-chloroethylamine hydrochloride at different ratios, producing materials with varying nitrogen contents [118]. Higher substitution levels resulted in reduced performance due to lignin's structural degradation. Adsorption tests conducted with Congo Red (CR) and Methylene Green (MG) (Fig. 2) revealed opposite pH-dependent behaviors: CR adsorption was favored under acidic conditions (maximum at pH 3) due to the dye's negative charge, whereas MG adsorption peaked at pH 9, corresponding to its cationic nature. Maximum removal exceeded 95% for both dyes. Reusability tests, performed using deionized water/ethanol (1:1) cycles, showed that MG maintained its performance, while CR adsorption decreased to 78% [118].

Softwood lignin was extracted through the Withstatter method, then it was converted into sorbents by microwave and acid precipitation methods. This was basically done to gain information about the influence of the treatment processes applied to lignin on the efficiency of adsorbance of a water soluble dye, Rhodamine B, which follows the previously described mechanisms (π - π -stacking interaction, hydrogen bonding, electrostatic adsorption) [119]. Both modified lignins outperformed raw lignin, showing faster equilibration and higher adsorption. The coarser morphology obtained after processing lignin, due to its chemical degradation, was considered one of the factors enhancing the lignin's adsorption capacity. The adsorption process was largely independent of pH in the range 2–10. Maximum RhB removal reached 86% for microwave-precipitated lignin and 96% for acid-precipitated lignin, confirming the superior effect of chemical modification [119].

A regenerated alkali lignin treated with deep eutectic solvents (DES) composed of choline chloride and oxalic acid (1:2 mol ratio) demonstrated strong adsorption toward Methylene Blue and Rhodamine B [120]. Optimal adsorption occurred at pH 8–9, attributed to electrostatic interactions with the positively charged dyes. Equilibrium was reached after 360 min for MB and 720 min for RhB, with adsorption capacities of 319.22 and 384.49 mg g^{-1} , respectively. At higher temperature (323K), capacities increased to 348.29 mg g^{-1} for MB and 551.05 mg g^{-1} for RhB. After four adsorption–desorption cycles, the material retained approximately 89% of its original capacity [120].

A biosorbent prepared by mechanical milling of α -chitin and lignin (1:1 ratio) in the presence of hydrogen peroxide exhibited effective adsorption of Direct Blue 71, achieving 9 mg g^{-1} at pH < 8.4 [121]. However, regeneration of the sorbent was not possible due to the strong binding of the dye, which could not be removed by the tested treatments [121].

Overall, lignin-based materials represent a sustainable and efficient option for dye removal from water. Their performance strongly depends on the chemical modification applied and the electrostatic nature of the target dye, requiring careful tuning of pH and surface functionalization to maximize adsorption.

4.1.4. Pharmaceuticals adsorption

Pharmaceutical residues, including antibiotics and anti-inflammatory drugs, are emerging pollutants of growing concern in aquatic systems. The functionalization of lignin with ionic or polar

groups enhances affinity also toward pharmaceutical molecules through hydrogen bonding and electrostatic interactions. Recent advances highlight the potential of both chemically modified lignins and hybrid lignin–biopolymer systems in effectively adsorbing drugs from water. The Fig. 6 displays the chemical structures of the active molecules discussed in this paragraph.

Magnetic lignin nanoparticles were synthesized for the adsorption of diclofenac by combining alkali lignin, FeCl₃, and sodium dodecyl sulfate, followed by freeze-drying and grinding [122]. The adsorption performance was evaluated across different pH values, with the highest capacity of 82.7 mg g^{-1} achieved at pH 5. At this pH, adsorption equilibrium was reached after 720 min. Reusability tests indicated a moderate decrease in performance after four cycles, with the capacity declining to 63.2 mg g^{-1} [122].

A biosorbent composed of lignin and chitin was prepared for the removal of non-steroidal anti-inflammatory drugs [123]. Adsorption tests conducted with ibuprofen and acetaminophen (paracetamol) revealed a higher affinity of ibuprofen for the sorbent, with maximum removal of 88% at pH 2, while acetaminophen reached 70% under the same conditions. Considering the higher efficiency of removal of ibuprofen shown by the sorbent, the authors attributed the higher performance to more favorable hydrophobic interaction with respect to other interaction mechanism such as hydrogen bonding and ion-dipole interactions. Adsorption efficiency decreased with increasing pH for both drugs. Regeneration experiments demonstrated that methanol and ethanol were effective desorbents, removing up to 80% of the adsorbed compounds [123].

Two classes of lignin-derived materials—crosslinked lignin and carboxymethylated crosslinked lignin—were synthesized and tested for the adsorption of ofloxacin, a fluoroquinolone antibiotic [124]. Crosslinking was performed with epichlorohydrin to obtain three materials of different crosslinking densities (LNE 1–3). From one of these (LNE-2), a series of carboxymethylated derivatives (LNEC 1–5) was produced using chloroacetic acid at varying degrees of substitution. Adsorption studies showed that the carboxymethylated samples exhibited higher adsorption capacities than the solely crosslinked ones. Within each family, adsorption increased with lower crosslinking density for LNE materials and with higher substitution degree for LNEC materials, reaching 0.8 mmol/g for LNEC-5 at pH 7. For both material types, optimal adsorption occurred at neutral pH [124]. The authors justified this behavior attributing the sorption mechanism to different intermolecular interactions, depending on the working pH: whilst at pH 2–4 or > 7 the

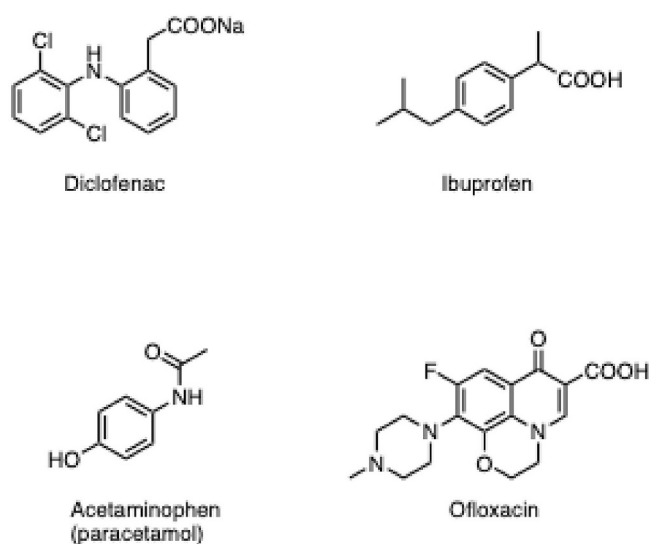


Fig. 6. Chemical structures of some of the pharmaceuticals currently considered emerging pollutants.

main interaction with ofloxacin was built on hydrogen bonding, at pH 4–7 a cooperative synergy between electrostatic interactions and π - π stacking, in which lignin works as a donor and the pollutant as an acceptor, enhances the adsorption efficiency.

Overall, the reported studies confirm the potential of lignin-based materials for pharmaceutical removal from aqueous media. Despite promising adsorption efficiencies and regeneration capabilities, further research is needed to expand their applicability across a wider range of pharmaceutical contaminants.

4.1.5. Other organic pollutants

Beyond metals, dyes, and pharmaceuticals, several studies have investigated lignin-based materials for the removal of other organic pollutants such as bisphenols, phenoxy acids, anilines, and nitro-aromatic compounds. The versatile aromatic structure of lignin, combined with its possible functionalization through grafting, crosslinking, or amination, enables strong π - π interactions, hydrogen bonding, and electrostatic attraction with a wide variety of organic molecules. An overview of the chemical structures of the emerging pollutants under consideration is shown in the Fig. 7.

Lignin-based aromatic porous polymers (LAPPs) were synthesized for the adsorption of bisphenol A (BPA) and bisphenol AF (BPAF) [125]. The polymers were obtained by polymerizing lignin with two different crosslinkers—1,4-dichloroxylylene and 4,4'-bis(chloromethyl)-1,1'-biphenyl using FeCl_3 as a catalyst in 1,2-dichloroethane, producing LAPP-1 and LAPP-2, respectively. Characterization revealed that LAPP-2 contained a higher density of phenolic rings, resulting in improved adsorption compared to LAPP-1. Adsorption capacities reached 349.42 and 375.69 mg g^{-1} for BPA, and 354.91 and 398.59 mg g^{-1} for BPAF, respectively. The materials maintained stable performance between pH 2–10, with a sharp decrease at pH 12, and showed minimal loss in adsorption capacity after five reuse cycles [125].

Bifunctional porous polyethyleneimine-grafted lignin microspheres were developed for the adsorption of 2,4-dichlorophenoxyacetic acid [126]. The microspheres were prepared via inverse suspension polymerization using epichlorohydrin as a crosslinker. The material demonstrated >90% adsorption efficiency across a wide pH range (4–10), reaching equilibrium within 90 min. Controlled release experiments showed pH-dependent behavior, with 57% of the adsorbed compound released at pH 2 after 72 h and 88% released at pH 11 after 24 h. After five adsorption–desorption cycles with anhydrous ethanol, the adsorption capacity decreased slightly from 95% to 88% [126].

Lignin grafted with acrylic acid was prepared for the removal of

aniline from aqueous solution [127]. The material was synthesized by dissolving lignin in methanol, adding acrylic acid, and initiating the reaction with ferrous ammonium sulfate and hydrogen peroxide. Adsorption performance was strongly pH-dependent, with the best results obtained at neutral pH (7), where hydrogen bonding was most favorable. The material adsorbed 72 mg g^{-1} of aniline, reaching equilibrium after 90 min. Regeneration tests using HCl showed remarkable stability, with only a 3.4% decrease in capacity after eight cycles (from 72 to 70 mg g^{-1}) [127].

Aminated lignin was synthesized for the adsorption of 2,4,6-trinitrotoluene (TNT) through a two-step process involving chlorination of lignin in 1,2-dichloroethane with AlCl_3 , followed by reaction with *N,N*-dimethylformamide and ethylenediamine [128]. The material exhibited an hypothesized hydrogen bonding interactions between the amino groups on lignin and the nitro-substituents on the aromatic ring of TNT and an adsorption capacity of 55.7 mg g^{-1} at 25 °C, with equilibrium reached after 24 h and higher adsorption at increased temperatures. Maximum adsorption occurred between pH 6 and 8. At higher pH, the adsorption efficiency was reduced by the formation of Janovski complexes of TNT, which dramatically changed the steric and electrostatic properties of the target pollutant. Reusability tests using ethanol and acetone as desorption solvents showed minimal losses after eight cycles, with capacities reduced to 95% and 92% of the initial values, respectively [128].

Lignin-based materials show strong potential for the removal of a wide range of organic pollutants from water. Their effectiveness largely depends on the type of chemical modification and the structure of the target molecules. However, further studies are needed to expand the spectrum of addressed contaminants and optimize regeneration strategies for practical application.

To provide a concise overview of the lignin-derived systems discussed, Table 3 summarizes the main sorbents and composite materials, their chemical modifications, and their adsorption or separation performance against various classes of pollutants. The examples highlight the versatility of lignin as a renewable feedstock for multifunctional materials capable of removing heavy metals, dyes, pharmaceuticals, and other organic contaminants through adsorption, membrane separation, or hybrid mechanisms.

5. Melanin and melanin-like materials

Melanins are a group of colored organic heteropolymers found in animals, plants, bacteria and fungi [131]. The understanding of the

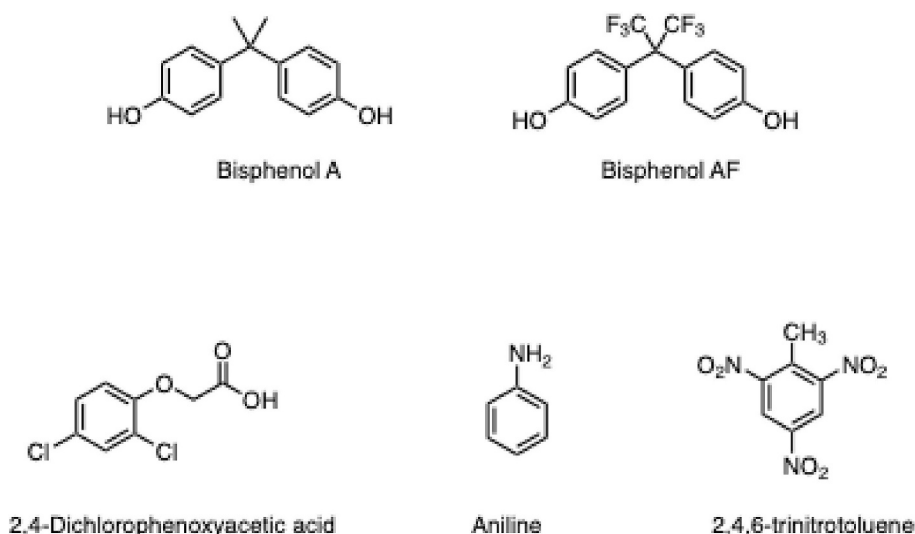


Fig. 7. Chemical structures of some of the emerging pollutants whose removal was explored using lignin-based sorbents.

Table 3

Lignin-based materials and their adsorption performance.

Material / System	Modification / Structure	Target pollutants	Removal efficiency / Capacity	Key features / Notes	Refs
Lignin–polypyrrole hydrogel	Cross-linked lignosulfonate + polypyrrole layer	Cu(II), Cr(VI), Co(II)	> 90%; salt below WHO limit	Stable at pH 1–14; 96% light absorption	[106]
Lignin–PU foam (PDA/PA)	Lignin–polyurethane with polydopamine + phytic acid	Cu(II); oils; dyes	Cu: 98 mg g ⁻¹ ; > 99% (oil); MB: 65%; RhB: 95%	Hydrophilic/oleophobic; reusable (10 cycles)	[107]
Lignin/silk fibroin membrane	Electrospun multilayer (lignin–PAN/silk)	Cu(II), Cd(II), dyes	Cu: 35%; Cd: 84.6% rejection	Hydrophobic surface; layered structure	[108]
Lignin nanotraps	Lignin + DETA + CS ₂ + formaldehyde	Ag(I), Pb(II), Cu(II), multi-ions	> 99% (multi-ion)	Optimum pH 5–7; equilibrium 30 min	[109]
Porous lignosulfonate–acrylic acid sorbent	Crosslinked with citric acid + MBA	Cu(II), Pb(II)	Cu: 250 mg g ⁻¹ ; Pb: 300 mg g ⁻¹	pH 7; equilibrium 60 min; 80% capacity after 5 cycles	[110]
Silica–lignin hybrid	Sol–gel composite	Co(II)	63% removal	Optimum pH ≥ 7; poor regeneration	[111]
Dual-modified lignin	Sequential reactions: formaldehyde/CS ₂ + chloroacetic acid	Pb(II)	~99% removal	Endothermic; 30% loss after 3 cycles	[112]
Quaternary ammonium–Zn (II) lignin sorbent	Lignin + 3-chloro-2-hydroxypropyl trimethylammonium chloride + ZnCl ₂	As(V)	72.6 mg g ⁻¹ (~70%)	pH 5; equilibrium 45 min; 11% loss after 5 cycles	[113]
Magnetic lignin–Fe ₃ O ₄ @SiO ₂ –NH ₂	Carboxymethylated lignin + Fe ₃ O ₄ core	Pb(II), Cu(II)	Pb: 90 mg g ⁻¹ ; Cu: 60 mg g ⁻¹	Equilibrium 30 s; acid instability	[114]
Lignin–PU foam	Acetylated lignin mixed with polyurethane	Oil/water separation	8 g g ⁻¹	Reusable (~90% after 50 cycles)	[115]
Lignin–PVA aerogel	Aminated lignin mixed with PVA and coated with methyl trimethoxy silane	Organic solvents	>90%	Hydrophobic at pH 1–14; reusable (10 cycles)	[116]
Lignin–PP sponge	PP sponge with various percentages of added lignin	Oils	>90%	Reusable: slight decrease in adsorption during the first three cycles, then stable at ~80% of the starting performance	[117]
Lignin–CuO–PDA mesh	PDA + Cu(OH) ₂ nanowires	Oil/water separation	> 99% efficiency	Hierarchical structure; gravity-driven separation	[129]
Amine–Lignin sorbent	Lignin crosslinked with 2-chloroethylamine	CR, MG	>95%	pH dependant for ionic dyes; reusable: MG adsorption stable, CR decreases to ~78%	[118]
Precipitated lignin sorbent	Microwave/acid precipitation of lignin	RhB	MW: 86%; acid: 96%	pH range: 2–10	[119]
Regenerated alkali lignin	Alkali lignin regenerated with DES	MB, RhB	MB: 319 mg g ⁻¹ , RhB: 38 mg g ⁻¹	Higher adsorption at higher temperature; reusable: ~89% after four cycles	[120]
Lignin–α-chitin sorbent	Milling in presence of hydrogen peroxide	DB 71	9 mg g ⁻¹	Not reusable: the dye binds too strongly to the sorbent	[121]
Lignin–P(E-alt-MA) hybrid gel	Poly (ethylene-alt-maleic anhydride) blend	Methylene blue (MB)	High, fast adsorption	Reusable ~3×; limited mechanical stability	[17]
Lignin–κ-carrageenan hydrogel beads	Natural melanin grafted κ-Ca hydrogel	MB, malachite green	MB: 48.6 mg g ⁻¹ ; MG: 37.8 mg g ⁻¹	Reusable (~50% after 3 cycles)	[130]
Magnetic lignin nanoparticles	Combination of alkali lignin, FeCl ₃ , and sodium dodecyl sulfate	Diclofenac	82.7 mg g ⁻¹	Best result at pH = 5; reusable: 63.2 mg g ⁻¹ after 4 cycles	[122]
Lignin–chitin sorbent	Alkali lignin mixed with chitin in presence of hydrogen peroxide	Ibuprofen, acetaminophen	Ibuprofen: 88%; acetaminophen: 70%	Best results at pH = 2; washing with MeOH or EtOH removes up to 80% of the drug	[123]
Cross-linked lignin sorbent	Lignin crosslinked with epichlorohydrin; with and without carboxymethylation	Ofloxacin	Up to 0.8 mmol/g	Optimal adsorption at pH = 7; carboxymethylated sorbents work better	[124]
LAPP-1 / LAPP-2 porous polymers	Lignin + FeCl ₃ catalyst + dichloroethylene or biphenyl crosslinker	Bisphenols (BPA, BPAF)	BPA: 349–375 mg g ⁻¹ ; BPAF: 354–398 mg g ⁻¹	Stable pH 2–10; reusable 5×	[125]
PEI-grafted lignin microspheres	Polyethyleneimine + epichlorohydrin crosslinking	2,4-dichlorophenoxyacetic acid	> 90% (pH 4–10)	Reusable (5×); pH-controlled release	[126]
Lignin–acrylic acid copolymer	Free-radical grafting (Fe ²⁺ /H ₂ O ₂ initiation)	Aniline	72 mg g ⁻¹ (pH 7)	3.4% loss after 8 cycles	[127]
Aminated lignin	Chlorinated lignin + ethylenediamine	2,4,6-trinitrotoluene (TNT)	55.7 mg g ⁻¹ (25 °C)	Max adsorption pH 6–8; stable after 8 cycles	[128]

chemical structure of melanin still needs clarification. Their isolation is usually difficult for their low abundance in biomasses, their insolubility and because they are chemically linked to other components of biological matrices as proteins, lipids, polysaccharides. Furthermore, their composition (qualitative and quantitative) is affected by biosynthesis conditions (e. g. origin, site of melanogenesis, reaction time, presence of L-cysteine or glutathione, metal ions etc.). Melanins are divided into five main classes, three of which contain nitrogen, i.e. eumelanin, neuromelanin and pheomelanin; the last two, pyromelanin and allomelanin,

are nitrogen free [132,133]. The Fig. 8 shows representative models of the chemical structures of the five classes of melanins.

Eumelanins typically contain 5,6-dihydroxyindole (DHI) and 5,6-dihydroxyindole-2-carboxylic acid (DHICA), that derive from dopaquinone, an oxidation product of L-DOPA. The ratio between carboxylated and non-carboxylated 5,6-dihydroxyindole relates to biosynthesis conditions, including the presence of divalent cations (Cu²⁺, Ni²⁺, Zn²⁺ and Co²⁺) [134,135]. DHICA oligomers display a non-planar twisting backbone with limited rotation freedom, and atropoisomerism arises at

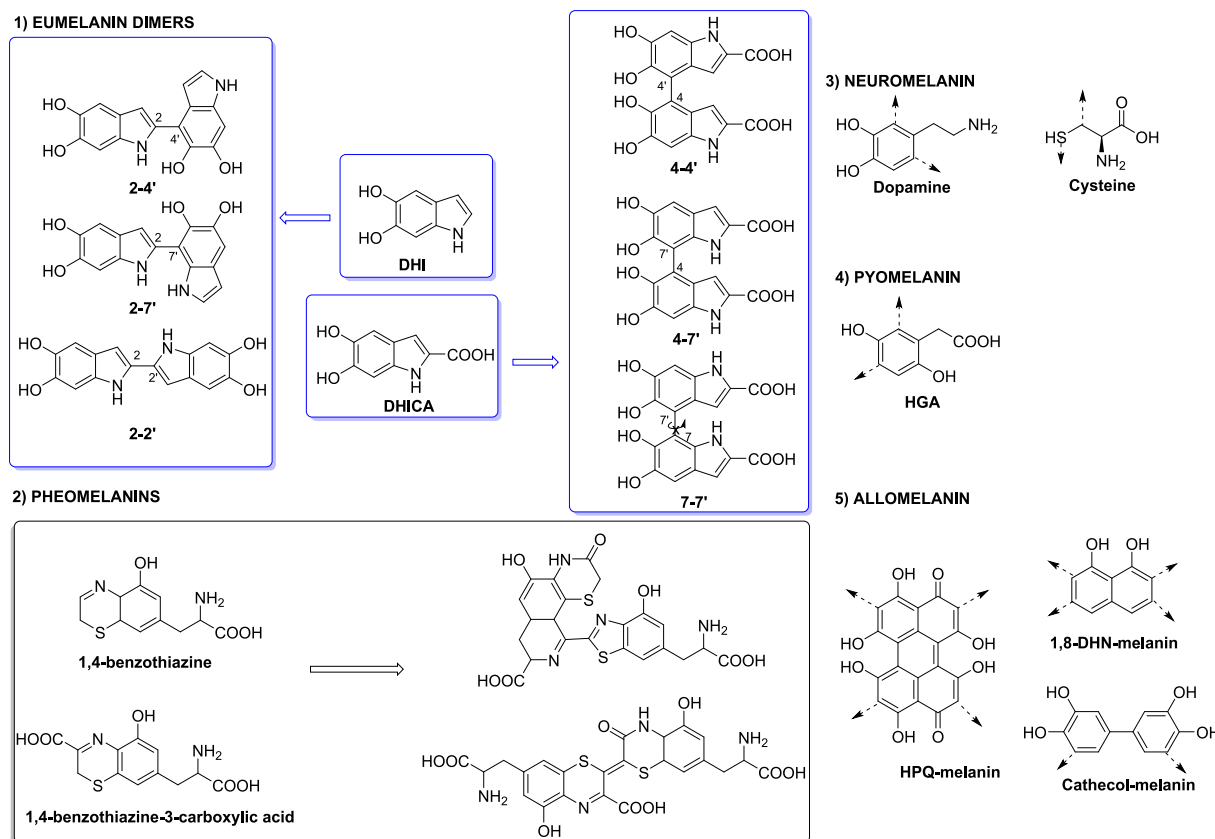


Fig. 8. Representative models of the chemical structures of the five different categories of melanin: eumelanin, pheomelanin, neuromelanin, and the two nitrogen free analogues, pyomelanin and allomelanin. The arrows indicate ideal prosecution of the conjugated structure by fusion with another monomer.

the interunit bonds [136]. Therefore, weak intermolecular interactions occur in DHICA polymer scaffolds, which are not amenable to π -stacking aggregation and show a stronger antioxidant behavior compared to their DHI counterpart, due to their less delocalized π -electron system. A further twist in DHICA polymer chain can be induced by the negative charge of deprotonated carboxyl groups [16].

Pheomelanins are sulfur-containing pigments, that dissolve at alkaline pH. Pheomelanins are formed by 1,4-benzothiazines and 1,4-benzothiazine-3-carboxylic acids in variable proportions. These building blocks derive from the reaction of L-cysteine addition to dopaquinone [137] and the following cyclization and oxidation. Pheomelanogenesis, with respect to eumelanogenesis, is favored by weakly acid pH [138]. Ion complexation, exposure to heat, to radiations or to free radicals have been able to explain partially the benzothiazines content occurring in pheomelanins, as a consequence of benzothiazines ring contraction [139].

Neuromelanin is also known as *substantia nigra* and derives from dopamine and cysteine dimers. Pyomelanin is a water soluble, nitrogen free, brown pigment and derives from polymerization of 2,5-dihydroxyphenylacetic acid (HGA) [140]. Allomelanin includes 1,8-dihydroxynaphthalene-melanin (DHN-melanin), 1,4,6,7,9,12-hexahydroxyperylene-3,10-quinone-melanin (HPQ-melanin) [141], and catechol-melanin [142].

Bacterial melanins display the highest composition uniformity because they are not affected by seasonal changes [143]. Unfortunately, bacteria strains cannot afford a large-scale production of melanins.

5.1. Melanin and melanin-like materials for water purification

Melanins possess a wide range of functional groups, including carboxyl, carbonyl, phenolic, hydroxyl, heterocyclic rings, and amine moieties, that provide numerous potential binding and adsorption/absorption sites for metal ions, organic molecules, and even biological

materials such as drugs. The results reported for melanin-based composites, together with their inherent biocompatibility, highlight melanin as a promising candidate for water bioremediation. This potential has also driven growing interest in melanin-like materials, such as polydopamine (PDA), which can overcome common limitations of adsorption technologies, including membrane fouling and the scarcity of reactive surface groups [144]. From a chemical perspective, PDA is a synthetic polymer formed by the oxidative self-polymerization of dopamine in alkaline aqueous solution, typically using tris(hydroxymethyl)aminomethane as an initiator [145]. According to the most accepted structural model, polydopamine does not exhibit a strictly linear or covalent framework but rather forms a supramolecular aggregate stabilized by hydrogen bonding, π - π stacking, and charge-transfer interactions. Its chemical and physical properties are strongly influenced by synthesis conditions, similarly to what is observed for natural melanins, with which PDA shares many structural and functional analogies [146]. PDA contains numerous catechol and amine residues that confer high surface reactivity and strong adhesion, even on wet substrates. Moreover, it can serve as a versatile platform for the covalent attachment of antifouling agents, such as polyethylene glycol or specific peptoids, thereby extending membrane lifetime and improving long-term stability [147].

Recent literature provides a comprehensive overview of PDA-polysaccharide hybrid systems, emphasizing their tunable surface chemistry, adsorption mechanisms, and sustainable potential for advanced water treatment [148].

A more recent review highlights how PDA-derived materials integrate adsorption, advanced oxidation, and membrane processes, demonstrating versatility in removing a broad range of pollutants [149].

5.1.1. Metals

Melanin and melanin-like materials have demonstrated significant

potential for heavy metal removal from aqueous environments.

Melanin extracted from squid ink sacs (20 mg/mL) effectively adsorbed Cr(VI) and Mn(II) ions (each at 2 mg/mL) from aqueous solutions at pH 4, achieving removal efficiencies greater than 90% within one hour. Compared to activated carbon at the same concentration, melanin showed superior adsorption capacity. The same study also reported the removal of *Vibrio parahaemolyticus* bacteria, achieving >90% efficiency after 24 h at melanin concentrations between 1 and 5 mg/mL [150].

Composite membranes incorporating fungal melanin particles from *Armillaria cepistipes* into polyurethane (PUR) and polycaprolactone (PCL) matrices were prepared by electrospinning for heavy metal removal [14]. The membranes were tested for adsorption of Pb(II), Cd(II), Ni(II), and Cr(III) from solutions with concentrations ranging from 25 to 600 µg/L at pH 6. Using 50 mg of membrane in 10 mL of solution, equilibrium was reached after 80 min. Melanin's negatively charged functional groups were responsible for metal adsorption, whereas PUR and PCL contributed primarily to the mechanical integrity of the composite.

Adsorption studies were performed in both single-ion and multi-ion systems to account for competition between metal ions with different affinities for melanin binding sites. In single-ion systems, adsorption efficiencies exceeded 90% for all metals, while in multi-ion systems, efficiency decreased in the order Cu(II) $\approx$ Pb(II) > Cr(III) > Ni(II) > Cd(II) > Zn(II) > Ca(II), consistent with their binding affinities [151]. Pb(II) displayed the highest affinity owing to its ability to coordinate with multiple melanin functional groups, including catechol, amine, and carboxyl moieties. In contrast, metals such as Cd(II), Zn(II), and Ca(II) bind primarily to carboxyl groups, as also demonstrated in melanized cationic cellulose nanofiber (C-CNF) composites [144].

Melanin nanoparticles coated onto hydrophobic polyvinylidene fluoride (PVDF) membranes further demonstrated efficient adsorption of mercury, chromium, lead, and copper ions [152]. The melanin coating enhanced the hydrophilicity of PVDF, improving surface interaction with metal ions. Maximum removal efficiencies of 87.6%, 88.5%, 91.8%, and 95.8% were achieved for mercury, chromium, lead, and copper, respectively, under batch conditions at 318 K and pH 3 (for chromium) or pH 5 (for other metals).

Additionally, polydopamine (PDA) membranes (200 $\pm$ 20 nm thick; 3 $\times$ 3 cm² surface area) coated on stainless steel exhibited selective electrochemical removal of Mg(II) from brines containing Li(I) and Mg(II) at concentrations of 350 µM and 100 µM, respectively [153]. The selective separation of Mg(II) was attributed to the reduction of *o*-quinone groups in PDA to catechol moieties, which selectively chelate Mg(II) ions, enabling the recovery of high-purity Li(I).

5.1.2. Oils and solvents

Melanin-inspired polymers, particularly polydopamine (PDA), have been widely employed in the fabrication of functional membranes for oil–water separation due to their strong adhesion, tunable wettability, and rich surface chemistry. These materials can be easily integrated into fibrous or metallic substrates, where they impart hydrophilicity and underwater oleophobicity, allowing water to permeate while effectively repelling oil droplets.

Nanofiber membranes with PDA clusters polymerized and embedded on the surface were fabricated via a solution blow-spinning process and demonstrated excellent oil–water separation performance, with efficiencies up to $\sim$ 99.7% under gravity and high permeation fluxes ($\sim$ 1550 $\frac{\text{L}}{\text{m}^2 \cdot \text{h}}$) across a range of oils. The flux increased further under mild pressure while maintaining separation efficiency. The rough surface morphology, combined with hydrophilic wettability and the embedded PDA domains, played a key role in repelling oil while enabling water transport [154].

In a related system, PDA was incorporated into a CuO nanowire-decorated mesh to form a hierarchical hybrid membrane capable of

gravity-driven oil–water separation. The ultrathin PDA coating introduced additional surface functionality and increased surface area, while the network of Cu(OH)₂ nanowires provided structural robustness and dual functionality, resulting in enhanced mechanical strength and separation efficiency [129].

Further developments have confirmed the effectiveness of PDA and other melanin-like coatings in such systems. Sulfonated PDA (SPD) coatings deposited in situ on PVDF nanofibers produced highly super-hydrophilic membranes (PVDF–SPD24h) able to sustain a remarkable water flux of 36,000 L m^{−2} h^{−1} and 99% oil–water separation efficiency. These membranes maintained 97.8% separation efficiency after 30 reuse cycles and exhibited excellent chemical durability, with less than 3 wt% mass loss after 48 h in acidic and alkaline media at 80 °C [155].

Similarly, PDA-assisted 2D polyethersulfone 2D/PES composite membranes have demonstrated outstanding oil–water emulsion separation performance with excellent antifouling behavior and chemical stability, combining fluxes exceeding 1000 $\frac{\text{L}}{\text{m}^2 \cdot \text{h}}$ with >99% separation efficiency [156].

Melanin-like and PDA-based coatings represent a sustainable and versatile approach to designing next-generation membranes for oil–water separation. Their unique combination of bioinspired chemistry, hierarchical surface structuring, and robust hydrophilicity enables exceptional flux, selectivity, and durability, making them promising candidates for industrial wastewater treatment and environmental remediation.

5.1.3. Dyes

Melanin and melanin-like materials, particularly polydopamine (PDA), have shown high efficiency in removing synthetic dyes from water due to their rich surface functionality and strong π – π and electrostatic interactions with aromatic dye molecules.

Electrostatic complexes formed between positively charged C-CNFs and negatively charged fungal melanin yielded foams with enhanced structural stability and compressive strength while maintaining high adsorption capacity. A foam prepared with 550 mg of fungal eumelanin per gram of C-CNFs removed up to 93% of crystal violet dye—even at ppm or ppb concentrations—within 180 min, and retained $\geq$ 80% efficiency after six adsorption–desorption cycles [157].

The derivatization of graphene hydrogels with PDA compensates for the low density of functional groups on pristine graphene, resulting in reusable adsorbents with high efficiency even after ten adsorption–desorption cycles. The composites can be regenerated using low-cost desorption agents such as 0.1 M HCl or ethanol, depending on the pollutant type. These PDA–graphene materials efficiently removed multiple contaminants, including Pb(II), Cd(II), Rhodamine B, and *p*-nitrophenol, through electrostatic attraction, chelation, and π – π interactions, showing significantly enhanced adsorption capacity compared to unmodified graphene. Adsorption experiments were performed in aqueous solutions with pH ranging from 3 to 6, where optimal removal occurred near neutrality. At pH values below 3, protonation of catechol and amine groups reduced binding affinity, whereas at pH > 7, the formation of metal hydroxides led to a decline in adsorption efficiency for both Rhodamine B and *p*-nitrophenol.

Sepia melanin was combined with poly(ethylene-*alt*-maleic anhydride) (P(E-*alt*-MA)) to produce a hybrid gel exhibiting faster and higher adsorption capacity for methylene blue compared to pure sepia melanin [17]. The catechol/quinone moieties in melanin provide active binding sites for cationic dyes, while the incorporation of P(E-*alt*-MA) enhances hydrophilicity by introducing additional carboxylate groups. The hybrid gel maintained adsorption performance across a wide pH range (2–12) and under high ionic strength conditions, with reusability up to approximately three cycles. However, mechanical instability limits its long-term reuse.

A natural melanin/ κ -carrageenan (κ -Ca) grafted hydrogel was also developed for the adsorption of cationic dyes methylene blue and

malachite green (Fig. 2), achieving maximum capacities of 48.6 and 37.8 mg g⁻¹, respectively. The hydrogel demonstrated good reproducibility but moderate reusability, retaining about 50% of its capacity after three extended cycles [130].

In another system, a recyclable polyacrylonitrile (PAN)–graphene oxide (GO)–PDA composite membrane exhibited dual adsorption functionality toward both heavy metal ions and organic dyes. The material achieved adsorption capacities of 62.9 mg g⁻¹ for Cu(II), 306.7 mg g⁻¹ for methylene blue (MB), and 339.6 mg g⁻¹ for eriochrome black T (EBT), an azoic sulfonated dye [158]. In this hybrid membrane, graphene oxide contributes to increased surface functionality and overall adsorption capacity, while PDA enhances binding affinity and facilitates interfacial integration between GO and the PAN support.

Overall, melanin-like materials, particularly PDA, significantly improve dye adsorption through multifunctional interactions, offering high removal efficiency, good recyclability, and tunable pH stability when incorporated into hybrid and composite systems.

5.1.4. Other organic pollutants

Polydopamine-polystyrene nanofibers mat (PDA-PS NFsM) was used to remove 18 different antibiotics simultaneously in environmental water. The binding sites such as phenolics, quinones, amines are efficient to adsorb antibiotics and the nanofibrous matrix gives large surface area and easy recovery. The efficacy obtained is ≥ 85% for each antibiotic reaching equilibrium in around 5 min. Limitation may be in regeneration efficiency over many cycles, in fact material reusability was achieved for ~10 cycles [159].

The interactions between pollutants and melanin-based or melanin-like polymers are influenced by several parameters, including the accessibility of binding sites, the oxidation state of functional groups, competition between ions for adsorption sites, and the pKa and pH of the polymeric system. These complex variables can strongly affect adsorption performance, and therefore further studies are needed to elucidate the mechanisms governing these processes in detail [160]. Building on these considerations, Table 4 provides an overview of representative melanin and melanin-like systems, detailing their structural

characteristics, pollutant types, and corresponding adsorption or separation efficiencies.

6. Biodegradability, environmental fate, and end-of-life considerations

Biodegradability is a defining advantage of bio-based materials; however, in the context of water treatment, it must be regarded as a tunable design parameter rather than an unconditional benefit. The biodegradability and environmental impact of cellulose-, lignin-, melanin-, and PDA-based materials involve a complex balance between environmental degradability, operational stability, and reusability. To provide a comparative overview of these aspects, Fig. 9 summarizes the relative performance of the different material classes based on a qualitative assessment of the literature discussed in this review.

While pristine cellulose, lignin, and natural melanin are inherently biodegradable, their transformation into functional adsorbents and membranes often requires chemical modification, crosslinking, nanostructuring, or hybridization, altering their degradation pathways, kinetics, and environmental fate. For practical water purification systems, materials are expected to remain structurally and chemically stable during operation, while biodegradation becomes relevant primarily at the end of service life. Consequently, an optimal balance between operational durability and post-use degradability is required.

For cellulose-based systems, biodegradation is primarily enzymatic, driven by cellulolytic microorganisms under soil, composting, or sludge conditions. However, increased crystallinity (as in cellulose nanocrystals), dense network formation in electrospun membranes, and surface functionalization (e.g., TEMPO oxidation, grafting of charged or chelating groups) significantly retard degradation rates by limiting enzyme accessibility. While this reduced biodegradability is advantageous for maintaining membrane integrity and adsorption performance during use, it necessitates consideration of controlled end-of-life scenarios. In this regard, nanocellulose materials exhibit a favorable compromise, offering high operational stability with the potential for biodegradation under appropriate disposal conditions.

Table 4
Melanin and melanin-like materials for water purification.

Material / System	Composition / Functionalisation	Target pollutants	Removal efficiency / Adsorption capacity	Key features / Notes	Ref
Natural squid melanin	Extracted from squid ink sacs (20 mg/mL)	Cr(VI), Mn(II); <i>Vibrio parahaemolyticus</i>	> 90% removal (1 h, pH 4); antibacterial >90% (24 h)	Outperforms activated carbon; multifunctional (metal + microbial removal)	[150]
Fungal melanin / PUR–PCL composite membrane	<i>Armillaria cepistipes</i> melanin in polyurethane / polycaprolactone	Pb(II), Cd(II), Ni(II), Cr (III)	> 90% (single ion); multi-ion selectivity (Cu ²⁺ > Pb ²⁺ > Cr ³⁺)	Equilibrium 80 min; catechol–metal complexation; mechanical stability	[14]
Melanin–C–CNF foam	Fungal eumelanin electrostatically complexed with cationic cellulose nanofibers	Crystal violet (CV)	93% (1st cycle); ≥ 80% (after 6 cycles)	Electrostatic adsorption; equilibrium 180 min; reusable and stable	[157]
Melanin-coated PVDF membrane	Melanin layer on PVDF nanofibers	Hg(II), Cr(III), Pb(II), Cu(II)	88–96% removal (pH 3–5, 318 K)	Enhanced hydrophilicity; chemical stability in acid/base media	[152]
PDA membrane on stainless steel	Polydopamine coating (200 nm)	Mg(II) from Li/Mg brine	4.6 μmol binding capacity; selective Li ⁺ recovery	Electrochemical separation via catechol–quinone redox sites	[153]
PDA–polystyrene nanofiber mat (PDA–PS NFsM)	Electrospun PDA–PS nanofibers	Antibiotics (18 types)	≥ 85% for all; equilibrium 5 min; reusable ≈ 10 cycles	Phenolic/quinone/amine binding; large surface area	[159]
PDA-assisted 2D/PES composite membrane	PDA + WS ₂ /MoS ₂ nanosheets in polyethersulfone	Oil-in-water emulsions	Flux $6360 \frac{L}{m^2 \cdot h \cdot bar}$; 99.7% oil rejection	High antifouling; durable 15 cycles; PDA improves adhesion & hydrophilicity	[156]
Sulfonated PDA–PVDF membrane	In-situ polymerized sulfonated PDA coating on PVDF	Oil–water mixtures	Flux $36,000 \frac{L}{m^2 \cdot h}$; 99% separation	97.8% efficiency after 30 cycles; stable at 80 °C (pH 1–14)	[155]
PDA-functionalised graphene hydrogel	Graphene hydrogel derivatised with PDA	Pb(II), Cd(II), RhB, <i>p</i> -nitrophenol	High adsorption; efficient over 10 cycles	Reusable; easy regeneration (HCl 0.1 M / EtOH)	[161]
Sepia melanin / P(E-alt-MA) hybrid gel	Sepia melanin blended with poly(ethylene-alt-maleic anhydride)	Methylene blue (MB)	High adsorption; reusable ≈ 3 cycles	pH 2–12; limited mechanical stability	[17]
Melanin–κ-carrageenan hydrogel beads	Natural melanin grafted on κ-carrageenan	Methylene blue, malachite green	MB 48.6 mg g ⁻¹ ; MG 37.8 mg g ⁻¹	Good reproducibility; 50% capacity after 3 cycles	[130]
PAN–GO–PDA composite membrane	Polyacrylonitrile + graphene oxide + PDA	Cu(II), MB, EBT dyes	Cu 62.9 mg g ⁻¹ ; MB 306.7 mg g ⁻¹ ; EBT 339.6 mg g ⁻¹	Dual adsorption (metal + dyes); high stability	[158]

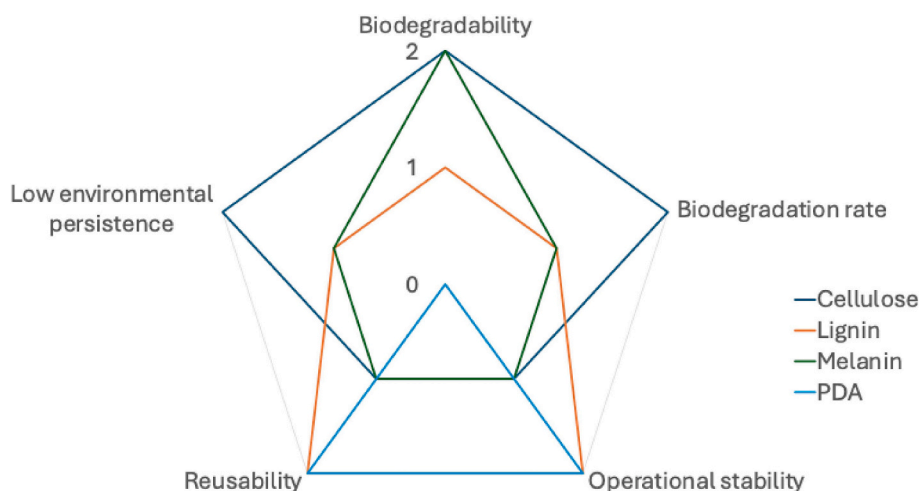


Fig. 9. Comparative sustainability-oriented framework for cellulose-, lignin-, melanin-, and polydopamine-based materials. The radar plot summarizes the relative performance of each material class in terms of biodegradability, biodegradation rate, operational stability, reusability, and environmental persistence. Scores on a 0–2 scale were assigned through a literature-informed qualitative assessment and represent an averaged evaluation of the studies discussed in this review. Higher scores indicate more favorable performance. The framework highlights the trade-offs frequently encountered in the design of bio-based water-treatment systems, where highly biodegradable materials may exhibit reduced long-term stability, while more persistent materials such as PDA often show improved durability and reusability. Reading code: 0 = poor / limited evidence; 1 = intermediate; 2 = favorable.

Lignin-based materials display inherently slower biodegradation due to their aromatic, heterogeneous structure and resistance to enzymatic attack. Chemical modification and crosslinking commonly employed to improve adsorption selectivity, mechanical robustness, and recyclability, further decrease microbial accessibility. As a result, lignin-based hydrogels, foams, aerogels, and membranes are best described as partially biodegradable, with degradation occurring over extended timescales. This intermediate persistence may be advantageous for long-term filtration applications, particularly where repeated regeneration is required, but it also underscores the importance of lifecycle assessment when lignin is proposed as a sustainable alternative to synthetic sorbents.

In the case of melanin and melanin-like materials, biodegradability is highly dependent on origin. Natural melanins produced by fungi or plants can undergo slow biological degradation, whereas synthetic analogues such as polydopamine (PDA) exhibit markedly higher environmental persistence. Although PDA is bio-inspired and offers exceptional metal chelation, antimicrobial activity, and antifouling behavior, its resistance to degradation challenges simplistic classifications as a “green” material. Therefore, melanin-based systems should be evaluated not only on adsorption performance but also on long-term environmental fate, particularly when employed as coatings or composite components in membranes.

Across all material classes, biodegradability is closely linked to regeneration and reuse strategies. Highly regenerable systems with long operational lifetimes reduce material throughput and environmental burden, even if they degrade slowly after disposal. Conversely, single-use or emergency water treatment materials may benefit from faster biodegradation once deployed. Future research should therefore focus on triggerable or programmable degradation mechanisms, such as enzyme-responsive linkages, pH-activated cleavage, or redox-sensitive crosslinks, enabling materials to remain stable during operation but degrade efficiently at end-of-life. Integrating biodegradability into the broader framework of performance, durability, and circularity is essential for the rational design of next-generation bio-based water purification technologies.

7. Limitations and challenges toward real-world implementation

The materials discussed in this review exhibit complementary

structural and functional properties, making them particularly promising for applications in water filtration technologies. Despite significant differences in availability, cellulose and lignin being the most abundant biopolymers on Earth, as well as in structural reproducibility, with lignin and melanin comprising highly complex chemical matrices, these materials share notable advantages. Their biodegradability and their ability to interact with pollutants through diverse mechanisms, like π – π stacking interactions enabled by the aromatic moieties in lignin and melanin, suggest that, in the future, it will be possible to design multifunctional adsorption systems based entirely on these bio-based materials. Such systems could achieve remarkable effectiveness in removing metal ions, organic dyes, and emerging organic pollutants. A visual summary of the key features and limitations of these three classes of materials is presented in Fig. 10.

As exemplified in Table 5, while a wide range of membranes obtained from the three classes of bio-based materials demonstrate high removal efficiencies under controlled laboratory conditions, most systems remain limited by short-term evaluation, lack of testing in complex matrices, and insufficient consideration of scalability and long-term operational stability.

Despite the extensive literature reporting promising performance of bio-based materials for water purification, several critical limitations continue to hinder their translation from laboratory-scale studies to real-world applications. A major issue lies in the predominance of simplified experimental conditions, including single-pollutant systems, synthetic aqueous solutions, and batch-mode evaluations. Real wastewater systems are inherently complex, containing a mixture of inorganic ions, natural organic matter, and multiple contaminants that can strongly interfere with adsorption processes. These simplified approaches therefore fail to capture the complexity of real wastewater matrices and continuous-flow operation. In addition, key aspects such as long-term stability, fouling behavior, and regeneration efficiency remain insufficiently investigated. Most studies report limited reusability over only a few cycles, without providing insight into material durability under prolonged use. Furthermore, the scalability and techno-economic feasibility of these systems are rarely addressed, particularly when complex functionalization strategies or energy-intensive synthesis routes are involved. While these limitations are common across all classes of bio-based materials, their specific nature and impact differ significantly among cellulose-, lignin-, and melanin-based systems, as discussed below.

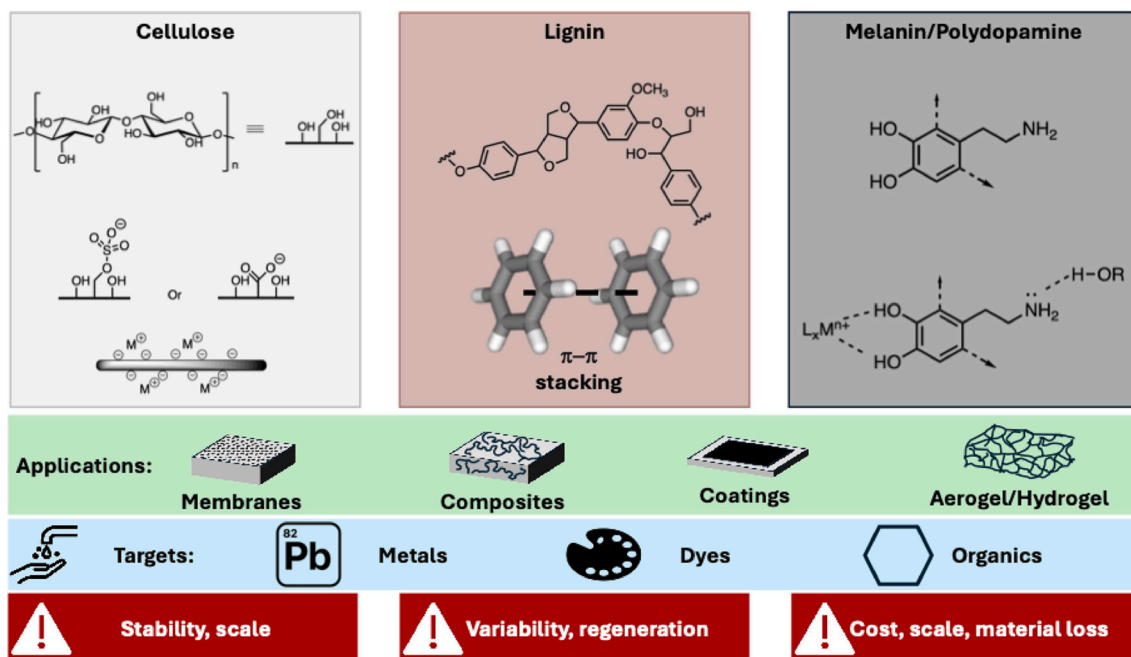


Fig. 10. A schematic summary of the contents of this work. A visual comparison between cellulose, lignin and melanin chemical structures. Main interactions involved in the adsorption process: electrostatic interactions, π - π stacking (for simplicity the figure shows only the face-to-face interaction), hydrogen bonding and chelation.

Table 5

Comparative performance and limitations of representative bio-based materials for water purification.

Material	Formulation	Pollutants	Performance	Conditions	Reusability	Limitations	Ref.
Nanocellulose	Membrane with TEMPO-CNF and non-woven mats	Cu (II)	Water permeance $>100,000 \text{ L m}^{-2} \text{ h}^{-1} \text{ MPa}^{-1}$ Absorption 64.3 mg g^{-1}	Pure water Continuous filtration	N/A	Recyclability not assessed. Working pH not explored	[83]
	Membrane with TEMPO-CNF/amyloid fibers	Methylene Blue, Cr (III), Fe(III), Ni(II), Cu(II)	Pore size compatible with rejection of molecules with diameter $> 3 \text{ nm}$	N/A	N/A	Pure TEMPO-oxidised nanocellulose membrane shows higher retention of metal ions than the composite	[89]
	Superhydrophobic CNF Aerogel	oils	$77\text{--}226 \text{ mg g}^{-1}$ depending on the organic liquid	ambient	~ 30 cycles	Recovery/scalability issues	[70]
	Electrospun membrane with silk fibroin	Water soluble dyes; Cu(II), Cd(II)	Retention rate $> 99.5\%$ for crystal violet, methylene blue, and brilliant green	N/A	N/A	Low metal rejection efficiency Susceptible to fouling	[108]
Lignin	Silica-lignin sol-gel composite	Co(II)	63% removal efficiency	$\text{pH} \geq 7$	poor	Loss after reuse due to acid regeneration. Loss of stability with temperature increase	[111]
	Nanotraps (decorated lignin nanoparticles)	Multi mono and divalent metals	$>99\%$	$\text{pH } 5\text{--}7$	limited data	Use of toxic chemicals for the synthesis	[109]
	Magnetic lignin nanoparticles	Pb(II)	90 mg g^{-1}	$\text{pH } 5$	N/A	Instability due to particles aggregation at acid pH	[114]
Melanin	Squid melanin	Cr (VI), Mn (II)	$>90\%$ from 2 mg/mL solution	$\text{pH } 4$	N/A	Low scalability	[150]
	Foam with cationic CNFs	Crystal violet	$\sim 93\%$; 80% after 6 cycles	N/A	≥ 6 cycles	Durability unknown	[157]
	PVDF-melanin hybrid membrane	Hg(II), Pb(II), Cu (II)	$88\text{--}96\%$	$\text{pH } 3\text{--}5$	not reported	Fouling not studied	[152]
PDA	Sulfonated PDA-PVDF Membrane	oil	$>99\%$, retained after 30 cycles	Continuous flow	30 cycles	Cost/scaling unclear	[155]
	PDA-graphene hydrogel	Pb(II), Cd(II), RhB, <i>p</i> -nitrophenol	$>90\%$	$\text{pH } 3\text{--}6$	~ 10 cycles	Easy regeneration, but complex chemical synthesis	[161]

7.1. Cellulose-based systems

Cellulose-based materials, particularly nanocelluloses, represent one of the most extensively studied classes of bio-based sorbents due to their high surface area, tunable surface chemistry, and excellent adsorption performance. However, several intrinsic limitations constrain their practical implementation. A key issue is the reliance on surface

functionalization to achieve high adsorption capacity, which often requires multi-step chemical modifications (e.g., TEMPO oxidation or grafting of charged groups). These treatments increase processing complexity and cost, and may also affect material stability, reducing crystallinity, altering morphology, or causing material loss during filtration.

From an application perspective, the majority of nanocellulose

systems are evaluated in batch mode, with limited translation to continuous-flow or membrane-based configurations where mass transport and pressure-driven processes play a critical role. As highlighted in Table 5, even highly permeable systems such as TEMPO-CNF-based membranes, which can achieve very high water fluxes, are typically tested under simplified conditions and often lack systematic evaluation of pH effects or long-term recyclability [83]. Similarly, comparative studies indicate that hybrid systems do not always outperform simpler formulations, as shown by the reduced metal-ion retention observed in TEMPO-CNF/amyloid fiber composites compared to pure nanocellulose membranes [89].

Mechanical stability also represents a significant challenge, particularly for highly porous structures such as aerogels and membranes, which may undergo structural collapse, compaction, or loss of active surface area during reuse. For example, superhydrophobic nanocellulose aerogels can exhibit good oil absorption capacity and reusability over several cycles, but still face limitations related to recovery and scalability [70]. Although some studies report recyclability over a limited number of cycles, systematic long-term durability assessments are still lacking.

In addition, issues related to material recovery in dispersed systems and the scalability of nanocellulose production, especially for bacterial cellulose, remain important barriers to large-scale implementation. These challenges are further compounded by the discrepancy between laboratory-scale evaluation and real wastewater conditions, where competing species and complex matrices can significantly affect adsorption performance.

A significant positive characteristic of nanocellulose membranes is their low fouling (accumulation of bacteria, colloids, salts, particles, other synthetic and proteins on the membrane surface) propensity mainly due to their hydrophilic characteristics. Surface charges on nanocellulose membranes repel similarly charged entities, thus reducing fouling tendency [162]. This tendency can be increased by common antifouling treatments, including zwitterionic and hydrophilic polymer coatings [163].

Finally, the excessive energy requirements of mechanical fibrillation methods significantly constrain the industrial-scale production of nanocellulose. At present, the production costs of cellulose nanomaterials are in the range of \$10 to \$50 per kilogram, while analysts predict that they should drop to \$1 to \$5 per kilogram for economically feasible operations [164]. Furthermore, no plant capable of producing nanocellulose at low cost, large scale, and in an environmentally friendly manner currently exists. These aspects further confirm that, beyond laboratory performance, economic feasibility and scalable manufacturing remain critical challenges for the practical deployment of nanocellulose-based systems. However, the valorisation of underutilized biomass feedstock such as agricultural waste residues, sawdust, and recycled cellulose fibers at low cost to obtain cellulose nanomaterials shows potential for pilot-scale commercialization.

Overall, while nanocellulose-based systems demonstrate excellent performance under controlled conditions, their practical application remains limited by the need for extensive functionalization, insufficient validation under realistic conditions, and challenges related to structural stability and scalability.

7.2. Lignin-based materials

The studies summarized in this review demonstrate the remarkable versatility of lignin-based materials for water purification, including hydrogels, aerogels, foams, membranes, nanoparticles and hybrid sorbents. However, a critical analysis reveals that the current literature focuses largely on describing adsorption capacities and removal efficiencies, whilst providing limited insight into the fundamental structure-property-performance relationships governing pollutant removal. Although improvements are often attributed to increased porosity, a larger surface area or the introduction of specific functional

groups, direct mechanistic evidence linking these material characteristics to adsorption kinetics, selectivity and long-term purification performance remains scarce. Consequently, comparisons between different lignin-derived architectures are often difficult, as adsorption experiments are carried out under highly variable conditions, pollutant concentrations and water chemistry.

The different classes of lignin-based materials exhibit different advantages and limitations:

- hydrogels and porous materials generally provide the highest adsorption capacities for heavy metals due to their large accessible surface area and abundant functional groups [106,110];
- aerogels and foams offer low density, high porosity and excellent performance in oil-water separation, often maintaining adsorption efficiencies of over 90% across multiple cycles [107,115,116];
- membrane systems provide filtration and adsorption capabilities, but often exhibit lower metal rejection efficiencies than dedicated adsorbents and may be more susceptible to fouling during prolonged operation [108];
- magnetic nanoparticles have rapid adsorption kinetics and easy recovery; however, their stability can be compromised under acidic conditions, limiting regeneration and reuse [114].

Despite the growing interest in lignin-based hybrid systems, the synergistic effects often cited are not always convincingly demonstrated. For example, the incorporation of polydopamine, silica, magnetic nanoparticles, polyurethane or polyvinyl alcohol clearly alters surface chemistry, wettability, porosity and mechanical properties [107,111,114,116], but quantitative comparisons with the corresponding single-component materials are rarely reported. Consequently, it remains difficult to determine whether performance improvements derive from genuine synergistic interactions or simply from the additive contribution of the individual components. Future studies should include systematic benchmarking against unmodified lignin and single-component reference materials, combined with advanced characterization techniques capable of correlating functional group density, pore architecture, surface charge and adsorption mechanisms with purification performance.

Furthermore, long-term stability has not yet been sufficiently investigated, as most studies evaluate performance over only a few adsorption-desorption cycles. Significant losses in adsorption capacity have been reported for several systems, including double-modified lignin-based sorbents [112], silica-lignin hybrids [111] and melanin-containing lignin composites [130], indicating progressive degradation of active sites or structural deterioration during regeneration. Regeneration itself remains a major limitation, particularly for systems exhibiting strong binding to pollutants, such as lignin-chitin-based adsorbents, where complete desorption is difficult to achieve [121]. The repeated use of acidic, alkaline or organic solvents for regeneration can further increase operating costs and reduce the material's life.

From an industrial perspective, scalability and commercialization remain unsolved. Although lignin is an abundant by-product of the pulp and paper industry, many adsorbents require multistep functionalization procedures, crosslinkers, or additional nanomaterials, which increase manufacturing complexity and production costs [12,103]. Furthermore, most studies are limited to laboratory-scale experiments using model pollutant solutions, while pilot-scale validation and continuous-flow testing remain unexplored. Environmental considerations have to be done, because lignin is biodegradable, but several hybrid systems incorporate components such as polydopamine (PDA), synthetic polymers, or inorganic nanoparticles, of which long-term environmental fate and degradation pathways remain poorly understood. Consequently, future sustainability assessments should consider not only adsorption efficiency but also material persistence and potential environmental impacts.

Future research should focus on defining correlations between

structure, properties and performance; developing standardized testing protocols; reducing the complexity of synthesis and production costs; and verifying the performance of materials under realistic operating conditions. In this way it can be possible transform lignin-based materials from promising laboratory-scale adsorbents into sustainable water purification technologies.

7.3. Melanin-based systems

Melanin and melanin-like materials, including polydopamine, are very promising platforms for water purification due to their rich surface chemistry and strong affinity toward both metal ions and organic pollutants. However, several challenges still limit their widespread application. A fundamental issue lies in the structural complexity and heterogeneity of melanin, which remains poorly defined at the molecular level and strongly dependent on its biological origin or biosynthesis conditions. The structural characterization of such a complex polymer often requires advanced solid-state spectroscopies, yet the chemical connectivity among monomeric units is still not fully understood. This limits the possibility of establishing clear structure–property–performance relationships.

An additional critical aspect that further complicates the interpretation of melanin-based systems is the relatively limited maturity of melanin chemistry compared to more established bio-based materials such as cellulose and lignin. While the latter have been extensively studied, melanin remains a poorly defined polymer, with significant gaps in the understanding of its molecular organization. In particular, the isolation of “pure” melanin is intrinsically challenging. Natural eumelanin is often co-extracted with inorganic residues or associated biopolymers such as chitin or proteins in biological matrices [165], which may contribute to the observed adsorption behavior. As a result, attributing specific properties exclusively to melanin is not always straightforward, although this aspect is often overlooked in the literature.

Scalability represents an additional critical limitation. Natural melanins are typically obtained in low yields and require complex extraction procedures, while synthetic analogues such as polydopamine involve multi-step chemical synthesis, increasing production costs and limiting large-scale applicability. As also highlighted in Table 5, melanin-based materials are rarely used as standalone systems and are more frequently incorporated into hybrid formulations, such as foams and membranes (e.g., melanin–CNF composites or PVDF–melanin systems) [152,157], indicating their dependence on supporting matrices to achieve adequate mechanical stability and processability. Furthermore, the lack of well-defined strategies for covalent integration into solid supports may lead to partial material release during operation, raising concerns about long-term stability.

Although high removal efficiencies are frequently reported—for example in the adsorption of transition metals or dyes using natural melanin or PDA-based systems [150,152,157]—performance is generally evaluated under simplified laboratory conditions, with limited validation in continuous-flow configurations or real wastewater matrices. Moreover, key aspects such as fouling behavior, long-term durability, and regeneration efficiency are not consistently assessed. As summarized in Table 5, reusability is often poorly reported or only evaluated over a limited number of cycles, while in several cases it is not assessed at all [150,152], making it difficult to determine long-term operational stability.

In addition, hybrid systems based on polydopamine (e.g., PDA-based membranes or PDA–graphene hydrogels) show promising multifunctional performance, but their synthesis complexity and scalability remain open challenges [155,161]. Despite providing a more controllable alternative to natural melanin, polydopamine is also an amorphous and insoluble material with a poorly defined structure, which may

incorporate residual components from the polymerization process, further complicating the establishment of clear structure–property–performance relationships.

Overall, these observations suggest that, despite promising adsorption performance, melanin-based systems remain at an early stage of technological maturity. The lack of systematic comparison with single-component systems, together with the limited availability of long-term and real-world data, represents a major barrier to their practical implementation in water treatment applications.

Overall, although all the investigated material classes display promising performance under laboratory conditions, significant differences remain in terms of technological maturity and implementation readiness. These aspects are summarized in Fig. 11, which provides a qualitative assessment of the current state of development of cellulose-, lignin-, melanin-, and PDA-based systems across different levels of validation and deployment and an interpretative framework about literature's maturity over the topic.

Overall, while bio-based materials demonstrate promising performance under controlled laboratory conditions, their practical deployment is still limited by insufficient evaluation under realistic conditions, lack of long-term stability data, and challenges related to scalability and process integration. As summarized in Table 5, these limitations manifest differently depending on the material class, highlighting the need for more standardized testing protocols and a stronger focus on structure–performance relationships to enable the transition toward real-world water treatment applications.

8. Conclusion

In conclusion, we can state that biomaterials like cellulose, lignin and melanin represent an interesting approach to water purification. Their environmental impact is limited (and, in some cases, positive) while also being good performers in these kinds of applications. The results reported in this review represent a good stepping stone to further explore the possibilities of these, and other, bio-based materials to employ in water purification. Despite the big progress that has already been made in the field, more studies are needed to expand the range of materials and their applications, especially in the elimination of organic compounds, such as drugs and dyes, from water.

The reviewed literature clearly demonstrates that bio-based materials such as cellulose, lignin, and melanin, along with their functionalized or composite derivatives, represent highly promising and sustainable alternatives for water purification technologies. Each class of biomaterial offers distinct advantages: nanocellulose provides tunable surface chemistry, mechanical robustness, and exceptional adsorption performance; lignin contributes aromatic and reactive functionalities for pollutant binding; while melanin and melanin-like polymers, such as polydopamine, combine versatile surface reactivity with biocompatibility and environmental resilience.

Functionalization and hybridization have proven essential to improving selectivity, adsorption kinetics, and recyclability across these systems. Modified nanocelluloses show high capacities for heavy metal removal and effective integration in filtration and electrospun membranes. Chemically modified lignins yield multifunctional sorbents and membranes capable of addressing diverse pollutants—ranging from metal ions to dyes, pharmaceuticals, and organic solvents—while melanin-based composites and PDA-coated systems demonstrate superior adsorption efficiencies (>90%) and long-term durability in both adsorption and oil–water separation applications.

Despite remarkable progress, several challenges remain before large-scale implementation can be achieved. These include improving regeneration and long-term stability, reducing fabrication costs, ensuring consistent performance under variable water chemistries, and integrating bio-based sorbents into scalable membrane and hybrid

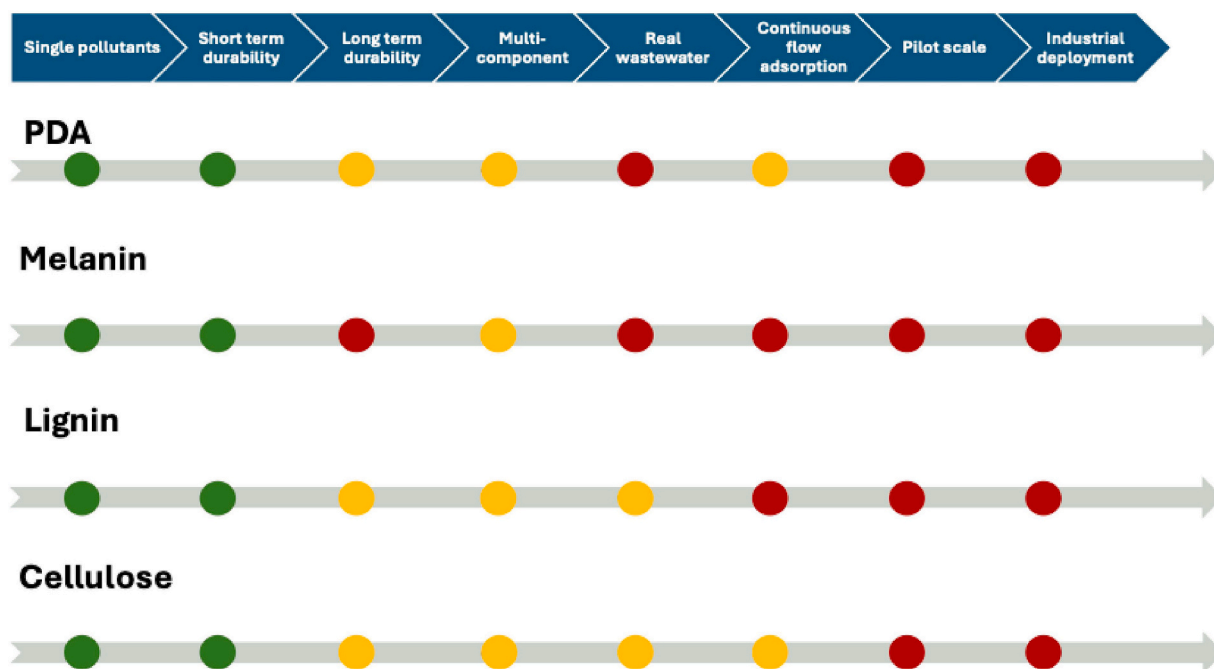


Fig. 11. Qualitative assessment of the maturity level of the literature on cellulose-, lignin-, melanin-, and polydopamine-based systems for water purification. The comparison considers the extent of available studies addressing single-pollutant systems, durability, multi-component mixtures, real wastewater validation, continuous-flow operation, pilot-scale demonstrations, and industrial implementation. The figure highlights the progressive decrease in available evidence when moving from laboratory-scale investigations toward technologically relevant conditions. While cellulose-based systems represent the most extensively studied class, significant gaps remain for all materials regarding long-term operation, pilot-scale validation, and industrial deployment. The assessment is intended as a qualitative interpretation of the literature discussed in this review rather than a quantitative technology-readiness evaluation. Colour code: Green = extensively investigated; Yellow = partially investigated; Red = limited or scarce evidence.

systems. Future research should focus on standardizing characterization protocols, elucidating adsorption mechanisms, and exploring synergistic effects in multi-component bio-composites. Furthermore, despite the significant progress reported for cellulose-, lignin-, and melanin-based materials, most of the studies discussed in this review are conducted under controlled laboratory conditions using simplified model systems. As a result, the reported adsorption capacities and removal efficiencies cannot be directly translated to real wastewater scenarios, where complex matrices, competing species, and variable conditions strongly affect performance. This highlights a critical gap between laboratory-scale research and practical application, emphasizing the need for future studies to evaluate these materials under realistic conditions and to assess their stability, reusability, and effectiveness in real wastewater systems.

Overall, cellulose-, lignin-, and melanin-derived materials provide a viable pathway toward eco-friendly, cost-effective, and circular water treatment technologies, aligning with global sustainability goals and offering realistic solutions for next-generation water purification. Future research should prioritize validation under real wastewater conditions, including multi-contaminant systems and long-term operation, to enable meaningful comparison across materials.

More broadly, the literature reviewed in this work indicates that the future development of bio-based water-treatment materials should move beyond the pursuit of high adsorption capacities alone. Material architecture, regeneration strategies, operational stability, and scalability emerge as equally important design criteria for real-world implementation. The conceptual frameworks proposed in this review further highlight that different bio-based materials offer distinct advantages and limitations, suggesting that future progress will likely rely on tailored formulations and hybrid approaches rather than on a single universal material platform.

CRediT authorship contribution statement

Federico Paolino: Writing – review & editing, Writing – original draft, Data curation, Conceptualization. **Asanda Mtibe:** Writing – review & editing, Writing – original draft, Conceptualization. **Nomvuyo Nomadolo:** Writing – review & editing, Writing – original draft, Data curation. **Rosarita D'Orsi:** Writing – review & editing, Writing – original draft, Supervision, Conceptualization. **Jeannette J. Lucejko:** Writing – review & editing, Writing – original draft, Supervision. **Alessandra Operamolla:** Writing – review & editing, Writing – original draft, Funding acquisition, Conceptualization. **Maya Jacob John:** Writing – review & editing, Writing – original draft, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Alessandra Operamolla reports financial support was provided by Government of Italy Ministry of Foreign Affairs and International Cooperation. Maya Jacob John reports financial support was provided by National Research Foundation. Federico Paolino reports financial support was provided by Ministry of University and Research. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work was supported by the Italian Ministry of Foreign Affairs and International Cooperation (MAECI), Grant Number PGR01179, CUP I53C22003350001 and by the National Research Foundation (NRF), South Africa, Grant Number 150552. F.P. thanks the Ministry of

University and Research (MUR) for the funding under D.M. 630/2024, CUP I51J24000290007.

Data availability

No data was used for the research described in the article.

References

- [1] L.E. Mofokeng, L. Hlekelele, J. Moma, Z.N. Tetana, V.P. Chauke, Energy-efficient CuO/TiO₂@GCN cellulose acetate-based membrane for concurrent filtration and Photodegradation of Ketoprofen in drinking and groundwater, *Appl. Sci. (Switz.)* 12 (2022).
- [2] L.E. Mofokeng, L. Hlekelele, Z.N. Tetana, J. Moma, V.P. Chauke, CuO-doped TiO₂ supported on graphitic carbon nitride for the Photodegradation of Ketoprofen in drinking and groundwater: process optimization and energy consumption evaluation, *ChemistrySelect* 7 (2022).
- [3] R.W. Herschy, Water Quality for Drinking: WHO Guidelines, in: *Encyclopedia of Earth Sciences Series*, 2012, pp. 876–883.
- [4] R. R. D. Thomas, E. Philip, S.A. Paul, A. Madhavan, R. Sindhu, P. Binod, A. Pugazhendhi, R. Sirohi, A. Tarafdar, A. Pandey, Potential of nanocellulose for wastewater treatment, *Chemosphere* 281 (2021) 130738.
- [5] R. Baby, B. Saifullah, M.Z. Hussein, Carbon nanomaterials for the treatment of heavy metal-contaminated water and environmental remediation, *Nanoscale Res. Lett.* 14 (2019).
- [6] D.G. Chandran, L. Muruganandam, R. Biswas, A review on adsorption of heavy metals from wastewater using carbon nanotube and graphene-based nanomaterials, *Environ. Sci. Pollut. Res.* 30 (2023) 110010–110046.
- [7] G. Li, R. Du, Z. Cao, C. Li, J. Xue, X. Ma, S. Wang, Research Progress in graphene-based adsorbents for wastewater treatment: preparation, adsorption properties and mechanisms for inorganic and organic pollutants, *C.J. Carbon Res.* 10 (2024).
- [8] M.S. Reza, S. Afroze, K. Kuterbekov, A. Kabyshev, K.Zh. Bekmyrza, M.N. Haque, S.N. Islam, M.A. Hossain, M. Hassan, H. Roy, Others, advanced applications of carbonaceous materials in sustainable water treatment, energy storage, and CO₂ capture: a comprehensive review, *Sustainability (Switzerland)* 15 (2023).
- [9] A.S. Norfarhana, R.A. Ilyas, N. Ngadi, A review of nanocellulose adsorptive membrane as multifunctional wastewater treatment, *Carbohydr. Polym.* 291 (2022) 119563.
- [10] T.C. Mokheba, E.R. Sadiku, M.J. Mochane, S.S. Ray, M.J. John, A. Mtibe, Mechanical properties of cellulose Nanofibril papers and their bionanocomposites: a review, *Carbohydr. Polym.* 273 (2021).
- [11] C.H.M. Camargos, L. Yang, J.C. Jackson, L.C. Tanganini, K.R. Francisco, S. R. Ceccato-Antonini, C.A. Rezende, A.F. Faria, Lignin and Nanolignin: next-generation sustainable materials for water treatment, *ACS Appl. Bio Mater.* 8 (2025) 2632–2673.
- [12] T. Wang, M. Jiang, X. Yu, N. Niu, L. Chen, Application of lignin adsorbent in wastewater treatment: a review, *Sep. Purif. Technol.* 302 (2022).
- [13] N. Kaushal, A.K. Singh, Advancement in utilization of bio-based materials including cellulose, lignin, chitosan for bio-inspired surface coatings with special wetting behavior: a review on fabrication and applications, *Int. J. Biol. Macromol.* 246 (2023).
- [14] A.N. Tran-Ly, J. Ribera, F.W.M.R. Schwarze, M. Brunelli, G. Fortunato, Fungal melanin-based electrospun membranes for heavy metal detoxification of water, *Sustain. Mater. Technol.* 23 (2020).
- [15] V. Lino, R. Castaldo, G. Gentile, P. Manini, Reusable melanin-based biosorbents for efficient methylene blue removal: the new frontier of Fungi-inspired Allomelanin coatings for sustainable water remediation processes, *Mater. Today Sustain.* 21 (2023).
- [16] E. Di Mauro, R. Xu, G. Soliveri, C. Santato, Natural melanin pigments and their interfaces with metal ions and oxides: emerging concepts and technologies, *MRS Commun.* 7 (2017) 141–151.
- [17] G. Panzarasa, A. Osypova, G. Consolati, F. Quasso, G. Soliveri, J. Ribera, F.W.M. R. Schwarze, Preparation of a Sepia melanin and poly(ethylene-alt-maleic anhydride) hybrid material as an adsorbent for water purification, *Nanomaterials* 8 (2018).
- [18] H. Voisin, L. Bergström, P. Liu, A. Mathew, Nanocellulose-based materials for water purification, *Nanomaterials* 7 (2017) 57.
- [19] M.N.F. Norrahim, H. Ariffin, T.A.T. Yasim-Anuar, M.A. Hassan, H. Nishida, T. Tsukegi, One-pot Nanofibrillation of cellulose and nanocomposite production in a twin-screw extruder, in: *Proceedings of the IOP Conference Series: Materials Science and Engineering*, 2018.
- [20] H. Ibrahim, N. Sazali, W.N.W. Salleh, A.F. Ismail, Nanocellulose-based materials and recent application for heavy metal removal, *Water Air Soil Pollut.* 232 (2021).
- [21] E.D. Smith, K.D. Hendren, J.V. Haag, E.J. Foster, S.M. Martin, Functionalized cellulose nanocrystal nanocomposite membranes with controlled interfacial transport for improved reverse osmosis performance, *Nanomaterials* 9 (2019) 125.
- [22] R. Amen, I. Elsayed, G.T. Schueneman, E.B. Hassan, Self-assembled Aminated and TEMPO cellulose nanofibers (am/TEMPO-CNF) aerogel for adsorptive removal of Oxytetracycline and chloramphenicol antibiotics from water, *Gels* 10 (2024) 77.
- [23] L. Dos Santos Silva, J. De Oliveira Carvalho, R.D. De Sousa Bezerra, M. Da Silva, F. Ferreira, J. Osajima, E. Da Silva Filho, Potential of cellulose functionalized with carboxylic acid as biosorbent for the removal of cationic dyes in aqueous solution, *Molecules* 23 (2018) 743.
- [24] O. Tkachenko, D. Diment, D. Rigo, M. Strømme, T.M. Budnyak, Unveiling the nature of lignin's interaction with molecules: a mechanistic understanding of adsorption of methylene blue dye, *Biomacromolecules* 25 (2024) 4292–4304.
- [25] B. Wang, Y.-C. Sun, R.-C. Sun, Fractional and structural characterization of lignin and its modification as biosorbents for efficient removal of chromium from wastewater: a review, *J. Leather Sci. Eng.* 1 (2019) 5.
- [26] M. Sorriaux, M. Sorieul, Y. Chen, Bio-based and robust Polydopamine coated Nanocellulose/amyloid composite aerogel for fast and wide-Spectrum water purification, *Polymers (Basel)* 13 (2021) 3442.
- [27] S. Ahsan, M.S. Rabbi, Recent advancement in Nanocellulose synthesis, Characterization and Application: A Review, *J. Polymer Sci. Eng.* 7 (2024) 4496.
- [28] J. Tang, J. Sisler, N. Grishkewich, K.C. Tam, Functionalization of cellulose nanocrystals for advanced applications, *J. Colloid Interface Sci.* 494 (2017) 397–409.
- [29] N. Rajendran, T. Runge, R.D. Bergman, P. Nepal, C. Houtman, Techno-economic analysis and life cycle assessment of cellulose nanocrystals production from wood pulp, *Bioresour. Technol.* 377 (2023).
- [30] W. Yu, L. Jiang, Application of carbon Fiber reinforced aluminum matrix composites in automotive industry, *Int. J. Automot. Manuf. Mater.* 3 (2024).
- [31] J. Cowie, E.M.T. Bilek, T.H. Wegner, J.A. Shatkin, Market projections of cellulose nanomaterial-enabled products - part 2: volume estimates, *TAPPI J.* 13 (2014) 57–69.
- [32] O.A. Battista, S. Coppick, J.A. Howsmon, F.F. Morehead, W.A. Sisson, Level-off degree of polymerization relation to Polyphase structure of cellulose fibers, *Ind. Eng. Chem.* 48 (1956) 333–335.
- [33] B.G. Rånby, Fibrous macromolecular systems. Cellulose and muscle. The colloidal properties of cellulose micelles, *Discuss. Faraday Soc.* 11 (1951) 158–164.
- [34] A. Mtibe, L.Z. Linganso, A.P. Mathew, K. Oksman, M.J. John, R.D. Anandjiwala, A comparative study on properties of Micro and Nanopapers produced from cellulose and cellulose Nanofibres, *Carbohydr. Polym.* 118 (2015) 1–8.
- [35] Y. Wang, H. Liu, Q. Wang, X. An, X. Ji, Z. Tian, S. Liu, G. Yang, Recent advances in sustainable preparation of cellulose nanocrystals via solid acid hydrolysis: a Mini-review, *Int. J. Biol. Macromol.* 253 (2023).
- [36] A. Operamolla, C. Mazzuca, L. Capodiceci, F. Di Benedetto, L. Severini, M. Titubante, A. Martinelli, V. Castelvetro, L. Micheli, Toward a reversible consolidation of paper materials using cellulose nanocrystals, *ACS Appl. Mater. Interfaces* 13 (2021) 44972–44982.
- [37] N. Lin, A. Dufresne, Surface chemistry, morphological analysis and properties of cellulose nanocrystals with gradiented sulfation degrees, *Nanoscale* 6 (2014) 5384–5393.
- [38] L. Spagnuolo, D. Beneventi, A. Dufresne, A. Operamolla, High yield synthesis of cellulose nanocrystals from Avicel by Mechano-enzymatic approach, *ChemistrySelect* 9 (2024).
- [39] A. Isogai, T. Saito, H. Fukuzumi, TEMPO-oxidized cellulose nanofibers, *Nanoscale* 3 (2011) 71–85.
- [40] R. D'Orsi, V.C. Canale, R. Cancelliere, O. Hassan Omar, C. Mazzuca, L. Micheli, A. Operamolla, Tailoring the chemical structure of cellulose nanocrystals by amine functionalization, *European, J. Organomet. Chem.* 26 (2023).
- [41] L. Spagnuolo, L. Micheli, A. Dufresne, D. Beneventi, A. Operamolla, Covalent lysozyme immobilization on enzymatic cellulose nanocrystals, *Chem. Eur. J.* 30 (2024).
- [42] R. D'Orsi, C. Danielli, M. Spennato, E. Guazzelli, E. Martinelli, F. Asaro, L. Gardossi, A. Operamolla, Cellulose nanocrystals and Rice husk surface functionalization induced by infrared thermal activation, *ChemSusChem* 18 (2025).
- [43] C.N. Nkuna, W. Mhike, V. Ojijo, M.B. Shongwe, M.J. John, Biocomposites and bionanocomposites from poly(Lactide) and cellulosic materials – a review, *Cellulose* 31 (2024) 4709–4732.
- [44] B.M. Tofanica, A. Mikhailidi, M.E. Fortună, R. Rotaru, O.C. Ungureanu, E. Ungureanu, Cellulose nanomaterials: characterization methods, isolation techniques, and strategies, *Crystals (Basel)* 15 (2025).
- [45] Z. Li, Y. Guan, N. Hu, J. Il Song, Prediction of elastic Modulus of cellulose nanofiber using density functional theory, *Mater. Chem. Phys.* 348 (2026).
- [46] T. Won, M.C. Goh, C. Lim, J. Moon, K. Lee, J. Park, K. Chung, Y. Kim, S. Lee, H. J. Hong, Others, recent Progress in cellulose Nanofibril hydrogels for biomedical applications, *Polymers (Basel)* 17 (2025).
- [47] R. Sharma, S.M. Jafari, S. Sharma, Antimicrobial bio-nanocomposites and their potential applications in food packaging, *Food Control* 112 (2020) 107086.
- [48] F. Barja, Bacterial Nanocellulose production and biomedical applications, *J. Biomed. Res.* 35 (2021) 310–317.
- [49] R. R. E. Philip, D. Thomas, A. Madhavan, R. Sindhu, P. Binod, S. Varjani, M. K. Awasthi, A. Pandey, Bacterial Nanocellulose: engineering, Production, and Applications, *Bioengineered* 12 (2021) 11463–11483.
- [50] D. Abol-Fotouh, M.A. Hassan, H. Shokry, A. Roig, M.S. Azab, A.E.H.B. Kashyout, Bacterial Nanocellulose from agro-industrial wastes: low-cost and enhanced production by Komagataeibacter Saccharivorans MD1, *Sci. Rep.* 10 (2020).
- [51] M. Ul-Islam, M.W. Ullah, S. Khan, J.K. Park, Production of bacterial cellulose from alternative cheap and waste resources: a step for cost reduction with positive environmental aspects, *Korean J. Chem. Eng.* 37 (2020) 925–937.
- [52] P.N. Baraskar, R.A. Samant, V.L. Gurav, Removal of heavy metal ions using Nano-cellulose Prepared from Rice husk: validation by differential pulse voltammetry, analytical, *Chem. Lett.* 13 (2023) 432–450.
- [53] R.T. Varghese, R.M. Cherian, T. Antony, A. Tharayil, H. Das, H. Kargazadeh, C. J. Chirayil, S. Thomas, A review on the best bioadsorbent membrane-

- Nanocellulose for effective removal of pollutants from aqueous solutions, *Carbohydr. Polym. Technol. Appl.* 3 (2022).
- [54] Y.J. Kim, J. Park, J. Bang, J. Kim, H.J. Jin, H.W. Kwak, Highly efficient Cr(VI) remediation by cationic functionalized Nanocellulose beads, *J. Hazard. Mater.* 426 (2022).
- [55] P.R. Sharma, A. Chattopadhyay, S.K. Sharma, L. Geng, N. Amiralian, D. Martin, B. S. Hsiao, Nanocellulose from spinifex as an effective adsorbent to remove cadmium(II) from water, *ACS Sustain. Chem. Eng.* 6 (2018) 3279–3290.
- [56] Q. Xu, Y. Wang, L. Jin, Y. Wang, M. Qin, Adsorption of Cu (II), Pb (II) and Cr (VI) from aqueous solutions using black wattle tannin-immobilized Nanocellulose, *J. Hazard. Mater.* 339 (2017) 91–99.
- [57] M. Kaur, J. Pal, Evaluation of efficiency of wheat straw Nanocellulose as Nanoadsorbent for the removal of divalent copper, Lead and zinc from aqueous solution, *Carbohydr. Polym. Technol. Appl.* 6 (2023).
- [58] J.N. Putro, S.P. Santos, S. Ismadi, Y.H. Ju, Investigation of heavy metal adsorption in binary system by Nanocrystalline cellulose – bentonite nanocomposite: improvement on extended Langmuir isotherm model, *Microporous Mesoporous Mater.* 246 (2017) 166–177.
- [59] T.S. Anirudhan, J.R. Deepa, J. Christa, Nanocellulose/Nanobentonite composite anchored with multi-carboxyl functional groups as an adsorbent for the effective removal of cobalt(II) from nuclear industry wastewater samples, *J. Colloid Interface Sci.* 467 (2016) 307–320.
- [60] T. Shahnaz, V. Sharma, S. Subbiah, S. Narayanasamy, Multivariate optimisation of Cr (VI), Co (III) and Cu (II) adsorption onto Nanobentonite incorporated Nanocellulose/chitosan aerogel using response surface methodology, *J. Water Process Eng.* 36 (2020).
- [61] J. Wei, Z. Yang, Y. Sun, C. Wang, J. Fan, G. Kang, R. Zhang, X. Dong, Y. Li, Nanocellulose-based magnetic hybrid aerogel for adsorption of heavy metal ions from water, *J. Mater. Sci.* 54 (2019) 6709–6718.
- [62] B. Geng, H. Wang, S. Wu, J. Ru, C. Tong, Y. Chen, H. Liu, S. Wu, X. Liu, Surface-tailored Nanocellulose aerogels with thiol-functional moieties for highly efficient and selective removal of Hg(II) ions from water, *ACS Sustain. Chem. Eng.* 5 (2017) 11715–11726.
- [63] B. Geng, Z. Xu, P. Liang, J. Zhang, P. Christie, H. Liu, S. Wu, X. Liu, Three-dimensional macroscopic Aminosilylated Nanocellulose aerogels as sustainable bio-adsorbents for the effective removal of heavy metal ions, *Int. J. Biol. Macromol.* 190 (2021) 170–177.
- [64] J. Li, S. Tan, Z. Xu, Anisotropic Nanocellulose aerogel loaded with modified UiO-66 as efficient adsorbent for heavy metal ions removal, *Nanomaterials* 10 (2020) 1–13.
- [65] L. Mo, H. Pang, Y. Lu, Z. Li, H. Kang, M. Wang, S. Zhang, J. Li, Wood-inspired Nanocellulose aerogel adsorbents with excellent selective pollutants capture, superfast adsorption, and easy regeneration, *J. Hazard. Mater.* 415 (2021).
- [66] J. Yang, Z. Luo, M. Wang, Novel fluorescent Nanocellulose hydrogel based on Nanocellulose and carbon dots for detection and removal of heavy metal ions in water, *Foods* 11 (2022).
- [67] S. Liu, Preparation of Nanocellulose grafted molecularly imprinted polymer for selective adsorption Pb(II) and Hg(II), *Chemosphere* 316 (2023).
- [68] Y.S. Tseng, R.R. Singhanian, A.C. Cheng, C.W. Chen, C. Di Dong, A.K. Patel, Removal of heavy metal vanadium from aqueous solution by Nanocellulose produced from Komagataeibacter Europaeus employing pineapple waste as carbon source, *Bioresour. Technol.* 369 (2023).
- [69] J. Sheetal, Pal, removal of Lead ions from aqueous solution by modified Nanocellulose, *Environ. Monit. Assess.* 196 (2024) 570.
- [70] Q. Shang, J. Chen, Y. Hu, X. Yang, L. Hu, C. Liu, X. Ren, Y. Zhou, Facile Fabrication of Superhydrophobic Cross-Linked Nanocellulose Aerogels for Oil–Water Separation, *Polymers (Basel)* 13 (2021) 625.
- [71] Y. Zhang, M. Yin, L. Li, B. Fan, Y. Liu, R. Li, X. Ren, T.-S. Huang, I.S. Kim, Construction of aerogels based on nanocrystalline cellulose and chitosan for high efficient oil/water separation and water disinfection, *Carbohydr. Polym.* 243 (2020) 116461.
- [72] P. Phanthong, P. Reubroycharoen, S. Kongparakul, C. Samart, Z. Wang, X. Hao, A. Abudula, G. Guan, Fabrication and evaluation of nanocellulose sponge for oil/water separation, *Carbohydr. Polym.* 190 (2018) 184–189.
- [73] J. Paul, S.S. Ahankari, Nanocellulose-based aerogels for water purification: a review, *Carbohydr. Polym.* 309 (2023) 120677.
- [74] N. Shaheed, S. Javanshir, M. Esmkhani, M.G. Dekamin, M.R. Naimi-Jamal, Synthesis of Nanocellulose aerogels and Cu-BTC/Nanocellulose aerogel composites for adsorption of organic dyes and heavy metal ions, *Sci. Rep.* 11 (2021).
- [75] N. Amiralian, M. Mustapic, M.S.A. Hossain, C. Wang, M. Konarova, J. Tang, J. Na, A. Khan, A. Rowan, Magnetic Nanocellulose: a potential material for removal of dye from water, *J. Hazard. Mater.* 394 (2020).
- [76] M.N. Thorat, A. Jagtap, S.G. Dastager, Fabrication of bacterial Nanocellulose/ Polyethyleneimine (PEI-BC) based cationic adsorbent for efficient removal of anionic dyes, *J. Polym. Res.* 28 (2021).
- [77] X. Zhang, I. Elsayed, C. Navarathna, G.T. Schueneman, E.B. Hassan, Biohybrid hydrogel and aerogel from self-assembled Nanocellulose and Nanochitin as a high-efficiency adsorbent for water purification, *ACS Appl. Mater. Interfaces* 11 (2019) 46714–46725.
- [78] J. Xue, E. Zhu, H. Zhu, D. Liu, H. Cai, C. Xiong, Q. Yang, Z. Shi, Dye adsorption performance of Nanocellulose beads with different carboxyl group content, *Cellulose* 30 (2023) 1623–1636.
- [79] J.X. Yap, C.P. Leo, D.J.C. Chan, N.H. Mohd Yasin, M.S. Sajab, *Chlorella vulgaris* Nanocellulose in hydrogel beads for dye removal, *Sep. Purif. Technol.* 324 (2023).
- [80] W. Maatar, S. Boufi, Microporous cationic Nanofibrillar cellulose aerogel as promising adsorbent of acid dyes, *Cellulose* 24 (2017) 1001–1015.
- [81] P. Jamwal, G.S. Chauhan, P. Kumar, B. Kumari, K. Kumar, S. Chauhan, A study in the synthesis of new Pinus Wallichiana derived spherical Nanocellulose hydrogel and its evaluation as malachite green adsorbent, *Sustain. Chem. Pharm.* 32 (2023).
- [82] B. Aoudi, Y. Boluk, M. Gamal El-Din, Recent advances and future perspective on nanocellulose-based materials in diverse water treatment applications, *Sci. Total Environ.* 843 (2022) 156903.
- [83] A. Mautner, Y. Kwaw, K. Weiland, M. Mvubu, A. Botha, M.J. John, A. Mtibe, G. Siqueira, A. Bismarck, Natural fibre-nanocellulose composite filters for the removal of heavy metal ions from water, *Ind. Crop. Prod.* 133 (2019) 325–332.
- [84] H. Gholami Derami, Q. Jiang, D. Ghim, S. Cao, Y.J. Chandar, J.J. Morrissey, Y.-S. Jun, S. Singamaneni, A robust and scalable Polydopamine/bacterial Nanocellulose hybrid membrane for efficient wastewater treatment, *ACS Appl. Nano Mater.* 2 (2019) 1092–1101.
- [85] Q. Zhu, Y. Wang, M. Li, K. Liu, C. Hu, K. Yan, G. Sun, D. Wang, Activable carboxylic acid functionalized crystalline Nanocellulose/PVA-co-PE composite Nanofibrous membrane with enhanced adsorption for heavy metal ions, *Sep. Purif. Technol.* 186 (2017) 70–77.
- [86] W. Zhu, M. Han, D. Kim, Y. Zhang, G. Kwon, J. You, C. Jia, J. Kim, Facile preparation of nanocellulose/Zn-MOF-based catalytic filter for water purification by oxidation process, *Environ. Res.* 205 (2022) 112417.
- [87] L. Valencia, S. Monti, S. Kumar, C. Zhu, P. Liu, S. Yu, A.P. Mathew, Nanocellulose/graphene oxide layered membranes: elucidating their behaviour during filtration of water and metal ions in real time, *Nanoscale* 11 (2019) 22413–22422.
- [88] Z. Wang, W. Zhang, J. Yu, L. Zhang, L. Liu, X. Zhou, C. Huang, Y. Fan, Preparation of Nanocellulose/filter paper (NC/FP) composite membranes for high-performance filtration, *Cellulose* 26 (2019) 1183–1194.
- [89] Y. Huang, P. Yang, F. Yang, C. Chang, Self-supported nanoporous lysozyme/nanocellulose membranes for multifunctional wastewater purification, *J. Membr. Sci.* 635 (2021) 119537.
- [90] H. Zhan, N. Peng, X. Lei, Y. Huang, D. Li, R. Tao, C. Chang, UV-induced self-cleanable TiO₂/Nanocellulose membrane for selective separation of oil/water emulsion, *Carbohydr. Polym.* 201 (2018) 464–470.
- [91] K. Peng, Y. Huang, N. Peng, C. Chang, Antibacterial Nanocellulose membranes coated with silver nanoparticles for oil/water emulsions separation, *Carbohydr. Polym.* 278 (2022).
- [92] Y. Huang, H. Zhan, D. Li, H. Tian, C. Chang, Tunicate cellulose nanocrystals modified commercial filter paper for efficient oil/water separation, *J. Membr. Sci.* 591 (2019).
- [93] H.Q. Zeng, H.Y. Hao, X. Wang, Z. Shao, Chitosan-based composite film adsorbents reinforced with Nanocellulose for removal of Cu(II) ion from wastewater: preparation, characterization, and adsorption mechanism, *Int. J. Biol. Macromol.* 213 (2022) 369–380.
- [94] H.F. Tan, B.S. Ooi, C.P. Leo, Future perspectives of Nanocellulose-based membrane for water treatment, *J. Water Process Eng.* 37 (2020).
- [95] D.A. Gopakumar, D. Pasquini, M.A. Henrique, L.C. De Moraes, Y. Grohens, S. Thomas, Meldrum's acid modified cellulose nanofiber-based Polyvinylidene fluoride microfiltration membrane for dye water treatment and nanoparticle removal, *ACS Sustain. Chem. Eng.* 5 (2017) 2026–2033.
- [96] J. Yan, T. Bai, Y. Yue, W. Cheng, L. Bai, D. Wang, J. Lu, M. Cao, S.Q. Shi, S. Huan, Others, nanostructured superior oil-adsorbent nanofiber composites using one-step electrospinning of Polyvinylidene fluoride/Nanocellulose, *Compos. Sci. Technol.* 224 (2022).
- [97] S. Rethinam, S.B. Kavukcu, T. Hemalatha, A.W. Aruni, A. Sendemir, C. Türkay, Cellulose based electrospun Nanofilters: perspectives on tannery effluent waste water treatment, *Cellulose* 29 (2022) 1969–1980.
- [98] R. Brandes, D. Belosinschi, F. Brouillette, B. Chabot, A new electrospun chitosan/phosphorylated Nanocellulose biosorbent for the removal of cadmium ions from aqueous solutions, *J. Environ. Chem. Eng.* 7 (2019).
- [99] H. Palacios Hiestroza, H. Urena-Saborio, F. Zurita, A.A. Guerrero de León, G. Sundaram, B. Sulbarán-Rangel, Nanocellulose and Polycaprolactone Nanospun composite membranes and their potential for the removal of pollutants from water, *Molecules* 25 (2020) 683.
- [100] D.S. Bajwa, G. Pourhashem, A.H. Ullah, S.G. Bajwa, A concise review of current lignin production, applications, products and their environment impact, *Ind. Crop. Prod.* 139 (2019).
- [101] R. D'Orsi, C.V. Irimia, J.J. Lucejko, B. Kahraman, Y. Kanbur, C. Yumusak, M. Bednorz, F. Babudri, M. Irimia-Vladu, A. Operamolla, Kraft lignin: from pulping waste to bio-based dielectric polymer for organic field-effect transistors, *Adv. Sustainable Syst.* 6 (2022).
- [102] R. D'Orsi, J.J. Lucejko, F. Babudri, A. Operamolla, Kumagawa and Soxhlet solvent fractionation of lignin: the impact on the chemical structure, *ACS Omega* 7 (2022) 25253–25264.
- [103] M. Sajjadi, F. Ahmadpoor, M. Nasrollahzadeh, H. Ghafari, Lignin-derived (Nano) materials for environmental pollution remediation: current challenges and future perspectives, *Int. J. Biol. Macromol.* 178 (2021) 394–423.
- [104] B.F. Mohazzab, B. Jaleh, M. Nasrollahzadeh, S. Khazalpour, M. Sajjadi, R. S. Varma, Upgraded valorization of biowaste: laser-assisted synthesis of Pd/calcium lignosulfonate nanocomposite for hydrogen storage and environmental remediation, *ACS Omega* 5 (2020) 5888–5899.
- [105] D. Han, Q. Hong, Emerging trends in cellulose and lignin-based nanomaterials for water treatment, *Int. J. Biol. Macromol.* 307 (2025) 141936.

- [106] S. Jiang, Z. Zhang, T. Zhou, S. Duan, Z. Yang, Y. Ju, C. Jia, X. Lu, F. Chen, Lignin hydrogel-based solar-driven evaporator for cost-effective and highly efficient water purification, *Desalination* 531 (2022) 115706.
- [107] J. Chen, J. Wu, Y. Zhong, X. Ma, W. Lv, H. Zhao, J. Zhu, N. Yan, Multifunctional superhydrophilic/underwater superoleophobic lignin-based polyurethane foam for highly efficient oil-water separation and water purification, *Sep. Purif. Technol.* 311 (2023) 123284.
- [108] Y. Zhang, S. Peng, X. Li, X. Wang, J. Jiang, X. Liu, L. Wang, Design and function of lignin/silk fibroin-based multilayer water purification membranes for dye adsorption, *Int. J. Biol. Macromol.* 253 (2023) 126863.
- [109] D. Xiao, W. Ding, J. Zhang, Y. Ge, Z. Wu, Z. Li, Fabrication of a versatile lignin-based nano-trap for heavy metal ion capture and bacterial inhibition, *Chem. Eng. J.* 358 (2019) 310–320.
- [110] X. Wang, X. Li, L. Peng, S. Han, C. Hao, C. Jiang, H. Wang, X. Fan, Effective removal of heavy metals from water using porous lignin-based adsorbents, *Chemosphere* 279 (2021) 130504.
- [111] T.M. Budnyak, J. Piątek, I.V. Pylypchuk, M. Klimpel, O. Sevastyanova, M. E. Lindström, V.M. Gun'ko, A. Slabon, Membrane-Filtered Kraft Lignin–Silica Hybrids as Bio-Based Sorbents for Cobalt(II) Ion Recycling, *ACS Omega* 5 (2020) 10847–10856.
- [112] Z. Zhang, C. Wu, Q. Ding, D. Yu, R. Li, Novel dual modified alkali lignin based adsorbent for the removal of Pb²⁺ in water, *Ind. Crop. Prod.* 173 (2021) 114100.
- [113] B. Zhang, W. Li, J. Ke, S. Fang, Effective adsorption of as(V) from aqueous solution by quaternary ammonium and Zn²⁺ decorated lignin-based sorbent, *Int. J. Biol. Macromol.* 261 (2024) 129883.
- [114] Y. Zhang, S. Ni, X. Wang, W. Zhang, L. Lagerquist, M. Qin, S. Willför, C. Xu, P. Fatehi, Ultrafast adsorption of heavy metal ions onto functionalized lignin-based hybrid magnetic nanoparticles, *Chem. Eng. J.* 372 (2019) 82–91.
- [115] U. Hwang, B. Lee, B. Oh, H.S. Shin, S.S. Lee, S.G. Kang, D. Kim, J. Park, S. Shin, J. Suhr, S.-H. Kim, J.-D. Nam, Hydrophobic lignin/polyurethane composite foam: An eco-friendly and easily reusable oil sorbent, *Eur. Polym. J.* 165 (2022) 110971.
- [116] Y. Yi, P. Liu, N. Zhang, M.E. Gibril, F. Kong, S. Wang, A high lignin-content, ultralight, and hydrophobic aerogel for oil-water separation: preparation and characterization, *J. Porous. Mater.* 28 (2021) 1881–1894.
- [117] A. Alasod, S.R. Islam, A. Farooq, G. Xu, Fabrication of polypropylene/lignin blend sponges via thermally induced phase separation for the removal of oil from contaminated water, *SN Appl. Sci.* 2 (2020) 1569.
- [118] D.H. Oh, J.W. Heo, Q. Xia, M.S. Kim, Y.S. Kim, Amine-crosslinked lignin for water pollution attributable to organic dye remediation: versatile adsorbent for selective dye removal and reusability, *Heliyon* 10 (2024) e37497.
- [119] A.O. Adeola, M. Cui, R. Naccache, Rhodamine B sequestration using acid-precipitated and microwave-treated softwood lignin: comparative isotherm, kinetics and thermodynamic studies, *Environ. Technol. Innov.* 32 (2023) 103419.
- [120] Y. Liao, W. Ge, M. Liu, W. Bi, C. Jin, D.D.Y. Chen, Eco-friendly regeneration of lignin with acidic deep eutectic solvent for adsorption of pollutant dyes for water cleanup, *Int. J. Biol. Macromol.* 260 (2024) 129677.
- [121] M. Wawrzkiwicz, P. Bartzak, T. Jesionowski, Enhanced removal of hazardous dye from aqueous solutions and real textile wastewater using bifunctional chitin/lignin biosorbent, *Int. J. Biol. Macromol.* 99 (2017) 754–764.
- [122] X. Ye, Y. Li, H. Lin, Y. Chen, M. Liu, Lignin-based magnetic nanoparticle adsorbent for diclofenac sodium removal: adsorption behavior and mechanisms, *J. Polym. Environ.* 29 (2021) 3401–3411.
- [123] S. Żółtowska-Aksamitowska, P. Bartzak, J. Zembruska, T. Jesionowski, Removal of hazardous non-steroidal anti-inflammatory drugs from aqueous solutions by biosorbent based on chitin and lignin, *Sci. Total Environ.* 612 (2018) 1223–1233.
- [124] B. Gao, P. Li, R. Yang, A. Li, H. Yang, Investigation of multiple adsorption mechanisms for efficient removal of ofloxacin from water using lignin-based adsorbents, *Sci. Rep.* 9 (2019) 637.
- [125] L. Sun, K. Chai, L. Zhou, D. Liao, H. Ji, One-pot fabrication of lignin-based aromatic porous polymers for efficient removal of bisphenol AF from water, *Int. J. Biol. Macromol.* 175 (2021) 396–405.
- [126] H. Wu, L. Gong, X. Zhang, F. He, Z. Li, Bifunctional porous polyethyleneimine-grafted lignin microspheres for efficient adsorption of 2,4-dichlorophenoxyacetic acid over a wide pH range and controlled release, *Chem. Eng. J.* 411 (2021) 128539.
- [127] X. Lin, J. Zhang, X. Luo, C. Zhang, Y. Zhou, Removal of aniline using lignin grafted acrylic acid from aqueous solution, *Chem. Eng. J.* 172 (2011) 856–863.
- [128] J. Zhang, X. Lin, X. Luo, C. Zhang, H. Zhu, A modified lignin adsorbent for the removal of 2,4,6-trinitrotoluene, *Chem. Eng. J.* 168 (2011) 1055–1063.
- [129] F. Zhang, Q. Zhao, D. Zhu, S. Gao, W. Fang, J. Jin, Multifunctional Polydopamine-modified CuO nanowire-haired membranes for efficient oil/water separation and dye removal, *Ind. Eng. Chem. Res.* 63 (2024) 9955–9963.
- [130] R. Hao, H. Ji, L. Gao, J. Chen, Y. Shi, J. Yang, C. Yuan, Grafted natural melanin κ -carrageenan hydrogel bead adsorbents: new strategy for bioremediation of cationic dye contamination in aqueous solutions, *Chem. Eng. Res. Des.* 199 (2023) 1–10.
- [131] V. Lino, P. Manini, Dihydroxynaphthalene-based Allomelanins: a source of inspiration for innovative technological materials, *ACS Omega* 7 (2022) 15308–15314.
- [132] W. Cao, X. Zhou, N.C. McCallum, Z. Hu, Q.Z. Ni, U. Kapoor, C.M. Heil, K.S. Cay, T. Zand, A.J. Mantanona, A. Jayaraman, A. Dhinojwala, D.D. Deheyn, M. D. Shawkey, M.D. Burkart, J.D. Rinehart, N.C. Gianneschi, Unraveling the structure and function of melanin through synthesis, *J. Am. Chem. Soc.* 143 (2021) 2622–2637.
- [133] P.M. Plonka, M. Grabacka, Melanin synthesis in microorganisms—biotechnological and medical aspects, *Acta Biochim. Pol.* 53 (2006) 429–443.
- [134] S. Ito, K. Wakamatsu, Chemistry of mixed Melanogenesis—pivotal roles of Dopachinone[†], *Photochem. Photobiol.* 84 (2008) 582–592.
- [135] A. Palumbo, M. Dischia, G. Misuraca, G. Protà, Effect of metal ions on the rearrangement of dopachrome, *Biochim. Biophys. Acta Gen. Subj.* 925 (1987) 203–209.
- [136] L. Panzella, G. Gentile, G. D'Errico, N.F. Della Vecchia, M.E. Errico, A. Napolitano, C. Carfagna, M. d'Ischia, Atypical structural and π -Electron features of a melanin polymer that Lead to superior free-radical-scavenging properties, *Angew. Chem. Int. Ed.* 52 (2013) 12684–12687.
- [137] S. Ito, A chemist's view of Melanogenesis, *Pigment Cell Res.* 16 (2003) 230–236.
- [138] K. Wakamatsu, A. Nagao, M. Watanabe, K. Nakao, S. Ito, Pheomelanogenesis is promoted at a weakly acidic <sc>pH</sc>, *Pigm. Cell Melanoma Res.* 30 (2017) 372–377.
- [139] S. Ito, K. Wakamatsu, T. Sarna, Photodegradation of eumelanin and Pheomelanin and its pathophysiological implications, *Photochem. Photobiol.* 94 (2018) 409–420.
- [140] S. Singh, S.B. Nimse, D.E. Mathew, A. Dhimmam, H. Sahastrabudhe, A. Gajjar, V. A. Ghadge, P. Kumar, P.B. Shinde, Microbial melanin: recent advances in biosynthesis, extraction, characterization, and applications, *Biotechnol. Adv.* 53 (2021) 107773.
- [141] N. Funai, M. Funabashi, Y. Ohnishi, S. Horinouchi, Biosynthesis of Hexahydroxypyrrolenequinone melanin via oxidative aryl coupling by cytochrome P-450 in *Streptomyces griseus*, *J. Bacteriol.* 187 (2005) 8149–8155.
- [142] F. Solano, Melanins: skin pigments and much more—types, structural models, biological functions, and formation routes, *New J. Sci.* 2014 (2014) 1–28.
- [143] A.N. Tran-Ly, C. Reyes, F.W.M.R. Schwarze, J. Ribera, Microbial production of melanin and its various applications, *World J. Microbiol. Biotechnol.* 36 (2020) 170.
- [144] N.H. Othman, N.H. Alias, N.S. Fuzil, F. Marpani, M.Z. Shahrudin, C.M. Chew, K. M. David Ng, W.J. Lau, A.F. Ismail, A Review on the Use of Membrane Technology Systems in Developing Countries, *Membranes (Basel)* 12 (2021) 30.
- [145] D.R. Dreyer, D.J. Miller, B.D. Freeman, D.R. Paul, C.W. Bielawski, Elucidating the structure of poly(dopamine), *Langmuir* 28 (2012) 6428–6435.
- [146] J. Liebscher, R. Mrówczyński, H.A. Scheidt, C. Filip, N.D. Hädäde, R. Turcu, A. Bende, S. Beck, Structure of Polydopamine: a never-ending story? *Langmuir* 29 (2013) 10539–10548.
- [147] A.R. Statz, R.J. Meagher, A.E. Barron, P.B. Messersmith, New Peptidomimetic polymers for antifouling surfaces, *J. Am. Chem. Soc.* 127 (2005) 7972–7973.
- [148] F. Liu, R. Jamal, T. Abdiryim, X. Liu, Polydopamine-based polysaccharide materials for water treatment, *Cellulose* 29 (2022) 8025–8064.
- [149] A. Savari, B.S. Tahir, A.M. Ahmed, R. Foroutan, B. Ramavandi, Application of polydopamine-based materials for advanced techniques in water and wastewater treatment, *Results Chem.* 15 (2025) 102256.
- [150] N.T. Le Na, P.T. Hoa, N.D. Thang, Natural Melanin as a Potential Biomaterial for Elimination of Heavy Metals and Bacteria from Aqueous Solution, 32 (n.d.).
- [151] J.-J. Oh, J.Y. Kim, Y.J. Kim, S. Kim, G.-H. Kim, Utilization of extracellular fungal melanin as an eco-friendly biosorbent for treatment of metal-contaminated effluents, *Chemosphere* 272 (2021) 129884.
- [152] V. Manirethan, N. Gupta, R.M. Balakrishnan, K. Raval, Batch and continuous studies on the removal of heavy metals from aqueous solution using biosynthesised melanin-coated PVDF membranes, *Environ. Sci. Pollut. Res.* 27 (2020) 24723–24737.
- [153] H. Park, Y.J. Kim, I.S. Kwon, L. Klosterman, C.J. Bettinger, Lithium purification from aqueous solutions using bioinspired redox-active melanin membranes, *Polym. Int.* 65 (2016) 1331–1338.
- [154] Z. Liu, Z. Xu, C. Liu, Y. Zhao, Q. Xia, M. Fang, X. Min, Z. Huang, Y. Liu, X. Wu, Polydopamine nanocluster embedded Nanofibrous membrane via blow spinning for separation of oil/water emulsions, *Molecules* 26 (2021) 3258.
- [155] V.D.C. Tinh, S.C. Bhandari, A. Bose, A sulfonated polydopamine coated-PVDF membrane development for oil/water separation via eco-friendly methodology, *J. Environ. Chem. Eng.* 11 (2023) 110560.
- [156] A.H. Jagaba, A.A. Elhussien, N. Baig, B. Salhi, N.A. Khan, I.H. Aljundi, A. M. Elsharif, I. Abdulazeez, Superhydrophilic polydopamine-assisted 2D/PES membranes for the effective separation of oil/water emulsions, *Sep. Purif. Technol.* 364 (2025) 132563.
- [157] A.N. Tran-Ly, K.J. De France, P. Rupper, F.W.M.R. Schwarze, C. Reyes, G. Nyström, G. Siqueira, J. Ribera, Melanized-cationic cellulose nanofiber foams for bioinspired removal of cationic dyes, *Biomacromolecules* 22 (2021) 4681–4690.
- [158] H. Wang, Z. Han, Y. Liu, M. Zheng, Z. Liu, W. Wang, Y. Fan, D. Han, L. Niu, Recyclable composite membrane of Polydopamine and graphene oxide-modified Polyacrylonitrile for organic dye molecule and heavy metal ion removal, *Membranes (Basel)* 12 (2022) 938.
- [159] S. Liang, H. Zhang, H. Dai, X. Wan, F. Zhu, Q. Xu, W. Ji, Efficient, rapid and simple adsorption method by polydopamine polystyrene nanofibers mat for removal of multi-class antibiotic residues in environmental water, *Chemosphere* 288 (2022) 132616.
- [160] L. Hong, J.D. Simon, Current understanding of the binding sites, capacity, affinity, and biological significance of metals in melanin, *J. Phys. Chem. B* 111 (2007) 7938–7947.
- [161] H. Gao, Y. Sun, J. Zhou, R. Xu, H. Duan, Mussel-inspired synthesis of Polydopamine-functionalized graphene hydrogel as reusable adsorbents for water purification, *ACS Appl. Mater. Interfaces* 5 (2013) 425–432.

- [162] S. Chen, L. Li, C. Zhao, J. Zheng, Surface hydration: principles and applications toward low-fouling/nonfouling biomaterials, *Polymer (Guildf)* 51 (2010) 5283–5293.
- [163] Y. Liang, H. Ma, A.A. Taha, B.S. Hsiao, High-flux anti-fouling nanofibrous composite ultrafiltration membranes containing negatively charged water channels, *J. Membr. Sci.* 612 (2020) 118382.
- [164] <https://eureka.patsnap.com/report-strategies-to-reduce-cost-of-nanocellulose-for-commodity-applications> (23/06/2026), (n.d.).
- [165] A.B. Mostert, S. Mattiello, S. Li, G. Perna, M. Lasalvia, P.F. Ambrico, J.V. Paulin, J. V.M. Lima, C.F.O. Graeff, J.W. Phua, M. Matta, A.J. Surman, R. Gunnella, M. Ambrico, Exploring the chemistry and composition of black soldier fly eumelanin, a material for a circular economy, *Mater. Adv.* 5 (2024) 8986–8999.