



# Advances in the modification and applications of cellulosic-based materials for wastewater remediation: a review

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## Abstract

Over the decades, the scarcity of fresh water has emerged as the most significant obstacles facing human societies. Many water sources have become threatened by contamination with different types of life-threatening pollutants, representing a major challenge to humans and leading to a shortage of getting safe drinking water. Therefore, many countries and researchers worldwide are trying to unite and overcome these challenges to identify innovative eco-friendly materials and techniques with considerable effectiveness for water purification. Recently, cellulose-based materials have shown considerable results in this field, which have attracted the attention of many scientists due to their unique and promising characterizations, which makes cellulosic material is an excellent substrate to develop attractive materials to treat wastewater. This review focuses on the new approaches in the modification and applications of biodegradable cellulose-based materials in treatment of wastewater and explains the advantages and disadvantages of such materials. On the other hand, this work highlighted the utilization of modified cellulose-based materials on the remediation of wastewater from different heavy metal ions (such as Cu(I,II), Pb(II), Tl, and F(− 1)), oil/water separation, removal of dyes, and removal of other organic pollutants. The review illustrated that cellulose-based materials are promising and effectiveness nanomaterials in its various forms to treat wastewater from different types of pollutants. Moreover, this work highlights the advantages and disadvantages of some applied characterization techniques in addition to the future challenges and prospect of value added of cellulosic-based materials for wastewater remediation.

**Keywords** Cellulose-based materials · Nanomaterials · Wastewater · Treatment

## Introduction

Natural water is vital for all living organisms, and clean water is demand for health and everyday life. Countless water resources have been contaminated by different types of pollutants such as dyes, metal ions, organic matter, oil, and other toxic species, and the issue represents a huge challenge nowadays and has become a major global concern (Rasaq et al. 2024; Haneen et al. 2023; da Paz Schiller et al. 2019; Junior et al. 2020). On the other hand, due to such contaminants, pure drinking water decreases over time in different regions worldwide, where such issues lead to various temporary or permanent significant threats to the ecosystem and ecology (Rasaq et al. 2024; Abdelhamid and Mathew 2021a, b). Therefore, the World Health Organization (WHO)

reported that most of the world's population (about 6.8 billion people) suffers from an acute shortage of drinking water, and so requires at least basic services and more efforts to reduce all types' pollutants and impurities to provide clean drinking water (Abdelhamid and Mathew 2021a, b). Globally, over 1.5 billion people worldwide live in countries suffer from water contamination, with more than 700 million people lacking access to clean waters. By 2050, about 87 countries worldwide projected to face water scarcity and more than 50% of the universal population will live in water-stressed regions. Currently, less than 3% of the total volumes of waters on Earth are fresh waters, 79% being glacial ice, and 20% being groundwater, in addition most fresh waters (3%) are not readily available to direct usage, and only 1.0% of these available amounts of freshwater is suitable to direct uses (Wang et al. 2021b, a; Abou-Shady et al. 2023; Khondoker et al. 2023; Shemer et al. 2023; Ren and

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Zhao 2024). On the other hand, many regions struggle with groundwater scarcity where about 35% of the Earth's land covered by arid areas (Ren and Zhao 2024). These unsuitable, limited, and insufficient available resources of freshwaters have high induce on the environment, ecosystems and human health. Many of domestic or regional conflicts, diseases, hunger, forced migration, and desertification may be attributed to the lack of reliable and safe fresh water (Shemer et al. 2023). Annually, due to the diseases associated with water pollutions and water shortages, millions of people are die (Wang et al. 2021b, a). Therefore, the scientific efforts have been focus on reduce pollution, protect, and conserve water sources to create a sustainable water infrastructure as an urgent need to find solutions to the problem of water scarcity. Recently, several techniques have been investigated to overcome these issues. Despite all these efforts, treating wastewater remains a more serious problem on an international scale (Thakur et al. 2022). Therefore, scientists direct their efforts to treating wastewater and maintaining a cleaner environment by removing or mitigating the toxicity of contaminations generated naturally or by human activities that could be inorganic (which is known as nondegradable, such as metal ions), organic (known as degradable, such as pharmaceutical compounds, antibiotics, oils, pesticides, soaps, fertilizers, petrochemicals, proteins, drugs, and dyes), or microorganism species (Rasaq et al. 2024; Sathish et al. 2024; Sawunyama et al. 2024; Ibrahim et al. 2024; Azimi et al. 2024). Generally, many factors determine the toxicity of water pollutants, such as concentrations, chemical forms, exposure time, and administration process (Rasaq et al. 2024). Moreover, the inadequate use of some materials for wastewater treatment affects human health and the entire ecosystem. Consequently, the need for eco-friendly and economical materials with considerable removal efficiencies has gained paramount importance (Azimi et al. 2024). To overcome the water scarcity problem and to acquire clean water from different sources of wastewater, numerous technologies and different types of promising materials/nanomaterials are currently being investigated as feasible and effective strategies. Recently, nanotechnology and nanomaterials have been used in different applications, such as the treatment of wastewater (Abdelhamid and Mathew 2021a, b). On the other hand, several types of nanomaterials have been studied and tested to use as effectiveness and promising materials with considerable toxin selectivity, a low dosage, and more economy in different wastewater treatment technologies such as flotation, filtration, membrane, separation, ion exchange, adsorption, chemical precipitation, etc. Therefore, nanotechnologies and nanomaterials offer valuable strategies for innovative wastewater treatment with promising applications such as compactness, minimal cost and low capital investment, simple activities, and accessibility over a different pollutant (Zhang et al. 2023a, b, c; Marimuthu et al. 2023). In

recent times, environmental biosorbents derived from have green materials gained attentions as eco-friendly biomaterials with promising properties for utilized in multidisciplinary fields due to their high efficiencies, low toxicities, high abundance, and low costs. On the other hand, the conventional adsorbent materials prepared from non-renewable resources have different environmental issues and resource depletion. Therefore, renewable adsorbents which are considered as sustainable alternative can offer a viable solutions to address these environmental concerns (Aslam et al. 2023). Also, many of common adsorbents present significant drawbacks such as its higher preparation costs, higher energy consumption, production of toxic sludge or other waste products, in addition to its limited recovery rates to regenerate. Therefore, utilization of green adsorbents derived from biowastes in treatment of wastewaters not only reduces the undesirable emissions, but also the economic feasibility of these technologies (Peng et al. 2020).

During the last decades, cellulose nanomaterials have high attention in various contaminated water applications due to their outstanding properties, such as biodegradability and renewability, sustainability, low carbon footprint, and availability in Earth's crust (Marimuthu et al. 2023). Recently, nanomaterials based on biomass attracted more attention due to their nontoxicity for the environment and humans. Researchers showed a great interest in biomass as an adsorbent due to its eco-friendliness, renewability, biocompatibility, and renderability. Cellulose is a linear polymer isolated from agriculture waste with molecular formula  $(C_6H_{10}O_5)_n$ . The cellulose has many OH groups (three for every glucose block), leading to highly dense intra and intermolecular H-bonds. Cellulose-based materials are sustainable, biodegradable, biocompatible, low-cost, and renewable. Cellulose and its derivatives are widely applied in numerous industries such as paper, clothes, food, beverage, biofuels, cosmetics, medicine, pharmaceutical, explosives, and other domains. Cellulose possesses adsorption limitations due to its low reactivity, removal rate, specificity, and reusability. Therefore, chemical modification is essential to improve its physical and adsorption characteristics. Carboxymethyl cellulose (CMC), derived from natural cellulose, is usually obtained in sodium salt form. The cellulose unit has three OH groups. Theoretically, these pendant hydroxyls can be easily transformed into the carboxylate groups. The modification is mainly located at the highest active  $C_6$  hydroxyl group. The presence of carboxylate groups with cellulose structure generates a negative charge on the surface, creating repulsion force among the cellulose molecules, and thus, the suspension stability of the CMC is enhanced with increasing substitution degree. Moreover, the presence of the groups offers the cellulosic materials benefit in the form of participation in complexation reactions with pollutant species.

In the current global trend towards sustainability and reducing production costs, various works were carried out using cellulosic nanomaterials. Where cellulose has clear properties over other materials that use chemicals in the synthesis process and may lead to environmental damage and also add extra costs to the production processes and pollutions remediation. Also, cellulose is naturally produced by plants, bacteria, fungi, and algae. Therefore, cellulose materials are eco-environmentally and with lower carbon footprint compared to those manufactured from petroleum products. Moreover, cellulose materials are characterized by special properties such as enhanced strength, flexibility, ease of functionalized, and biological compatibility. Various works conducted by researchers in the field of cellulose that targets industrial needs are due to the distinctive properties of cellulose materials such as the presence in large quantities production of cellulosic materials every year (zero cost), which ultimately leads to reducing the final cost of production, which is the most important factor for achieving economic success. To extend the application capacity of cellulose materials in various fields, textiles, biosensors, packaging materials electrical devices, and biomedical application, functional groups may be added or it may be integrated with other materials. Moreover, the advanced technology leads to innovated a novel cellulosic-based materials which can be used in different fields.

Based on the urgent need for sustainable and considerable effective materials to treat wastewater and decrease energy exhaustion, it is of high concern for sustainable development to investigate advanced eco-friendly materials of nature renewable sources which can be offer excellent inspiration to invent such materials. Due to its low cost, nontoxicity, renewable source, vast availability, eco-friendly nature, biodegradable and biocompatible, most abundant and renewable biopolymer on earth, in addition to thermal and chemical stable nature (Tang et al. 2024), cellulose is one of the most promising materials that can be used in various applications. Furthermore, due to its abundant and accessible ( $-OH$ ) groups which access to ease chemical functionalization the function groups to amine groups, carboxylic acid, thiol, and etc., cellulose can be obtained with various modified cellulose-based materials, like membranes, aerogels, adhesives, hydrogels etc., by various chemical or physical functionalization methods (Tang et al. 2024; Marimuthu et al. 2023). Therefore, and due to those excellent features, excellent mechanical, chemical, and optical properties, cellulose-based materials as promising materials have attracted a lot of attentions from the scientists as well as industries for the technologies of purification of wastewater in conjunction with sustainable development (Sinha et al. 2023; Marimuthu et al. 2023). Generally, nanocellulose could be classified into four classes: electrospun cellulose (ECCs), bacterial nanocellulose (BNCs), cellulose nanofibrils (CNFs), and

cellulose nanocrystals (CNCs) (Yu et al. 2021b, a). Cellulose nanofibrils (CNFs) and cellulose nanocrystals (CNCs) are the main two types of cellulose nanomaterials that have been eminently investigated in the wastewaters treatment (Marimuthu et al. 2023).

During the last decades, (CNFs) and (CNCs), which jointly referred to as cellulosic nanomaterials (CNs) and considered as sustainable nanomaterials, have been explored to apply on large scale in treatment technologies of wastewaters (Mohammed et al. 2018). (CNFs) are produced by either chemical or mechanical treatment while (CNCs) are obtained by chemically treatment and are differences mainly in their crystallinity and dimensions (Grishkewich et al. 2017). CNCs can be extracted from raw materials using biological, chemical, or physical pretreatment with similar chemical compositions as plant cellulose with rod-shaped nanomaterials (Zhang et al. 2023a, b, c). CNCs have amorphous regions and consist of crystalline cellulose fibers and possessed lots of desirable chemical and physical characteristics which offer a facile platform for chemical modifications (Grishkewich et al. 2017; Zhang et al. 2023a, b, c) so that these promising properties of CNCs make them useful building blocks in different critical applications.

Nanofibrillated cellulose materials (NFCs) have typical lengths of several micrometers and diameters of nanoscales. NFCs were first isolated by a Gaulin laboratory homogenizer from more than 40 years (Herrick et al. 1983; Turbak et al. 1983). As reported in the literature, the nanofibrillated cellulosic materials displayed two major drawbacks that affect their intrinsic physical characteristics which restricts its application in various fields. These limitations can be summarized in the higher hydrophilicities of these materials and large numbers of  $-OH$  groups (Missoum et al. 2013). Chemical surface modification processes are the most feasible solutions to overcome these limitations (Missoum et al. 2013). Therefore, surface modifications of nanofibrillated cellulose were investigated and gained great attentions from the scientists to improve its properties and to open the ways toward heavy and favorable works with wide areas of advanced applications (Kalia et al. 2014).

Cellulose nanocrystals (CNCs) belong to environmentally and green wastewater treatment material. CNC is typically highly crystalline needle-shaped fibrils prepared by acid hydrolysis of cellulosic precursor substance with dimensions ranging from 100 nm to several micrometers. CNC has a large density of available ( $OH$ ) groups on the CNC surface, which produces high repulsion force and enhanced hydrophilicity. Moreover, these actives of the  $OH$  groups promote interaction with several chemical species to customize the properties of the surface for different applications.

Bacterial cellulose (BC) has similar chemical structure to that of plants. Some bacterial strains produce the BC through the process of fermentation, consisting of D-glucose,

peptone, yeast extract, sodium dihydrogen phosphate, and citric acid. The backbone of the BC blocks enriches with OH groups interacting within H-bonding. This led to lining up the cellulose chain parallel into a crystalline structure. However, the obtained BC is free from lignocellulose, hemicellulose, pectin, and other bio-components; thus, it possesses higher crystallinity than other cellulosic resources. Moreover, BC is built of fibers that are entangled with each other in a 3D network membrane. BC's unique structure significantly improves their properties such as chemical stability, flexibility, tensile strength, high porosity, biocompatibility, high permeability, swellability, low toxicity, and durability.

Cellulose acetate (CA), a polymer derived from cellulose, widely is used in film-forming applications with excellent high hydrophilicity, pore structure network, biodegradability, and biocompatibility. CA membrane can be synthesized by various techniques as electrospinning, casting, and phase inversion. The electrospinning method of the CA produced the nanofiber membrane with specific molecular weight, possibly generating various layers of CA nanofibers and forming a continuous porous network structure. The CA membrane synthesized by the phase inversion usually possesses a dense structure with a porous sublayer, which leads to reduced flux.

## Cellulose-based materials and heavy metals

The rapid expansion of urbanization and industrialization process is accompanied by the discharge of serious organic and inorganic pollutants into water resources (Sun et al. 2022; Wang et al. 2022a, b; Liu et al. 2022a, b, c; Gao et al. 2022; Maaloul et al. 2021). Recently, the heavy metals dose in the whole ecosystem and human bodies has been raised because of quick growth in industrialization; the contamination by such types of pollutants has gained widespread concerns and paid more and more attention (Wang et al. 2022a, b; Liu et al. 2022a, b, c; Gao et al. 2022; Maaloul et al. 2021; Jawhid et al. 2021; Ai et al. 2022; Liu et al. 2022a, b, c; Song et al. 2022; Xing et al. 2021; Zeng et al. 2023; Elias et al. 2022). Heavy metal ions are typically discharged from mining, textile, metallurgy, electrical, non-ferrous metal smelting, and other industrial production. Heavy metals such as  $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ ,  $\text{Hg}^+$ ,  $\text{Cd}^{2+}$ ,  $\text{Cr}^{6+}$ ,  $\text{Co}^{2+}$ ,  $\text{Cs}^+$ , etc. can contaminate the food chain, damaging human and animal health. For example, the presence of  $\text{Pb}^{2+} > 0.07 \text{ mg/L}$  in fresh water causes human harm like kidney gastrointestinal bleeding, failure, and brain injury. Moreover, the concentration of  $\text{Cd}^{2+} > 0.005 \text{ mg/L}$  results in disorders of protein metabolism, lung, liver, kidney damage, infertility, and even cancer. Therefore, it seems necessary to develop materials and techniques for the remediation of the targeted heavy metals (Jawhid et al. 2021). Recently, several studies have

been developed to investigate considerable safe and efficient cellulose-based materials for removing inorganic pollutants, as illustrated in Table 1 [14–46].

## For Cu

Copper is a main trace species inside the human body, shared in the growth and metabolism processes. On the one hand, high doses of copper are aggregated in the human body and lead to numerous health diseases such as vomiting, diarrhea, headaches, and in severe cases, kidney failure. (Sun et al. 2022). On the other hand, severe complications such as pneumonia, tremors, and diabetes can occur as a result of drinking chronic ingestion of  $\text{Cu(II)}$ -contaminated water (Wang et al. 2022a, b).

A novel inorganic–organic biosorbent was investigated by Sun et al. (2022). The biosorbent was prepared via nanocellulose crosslinking with magnetic bentonite and modified with polyethyleneimine (PEI). The prepared PEI-grafted nanocellulose/magnetic bentonite composite (PNMBC) showed considerable potential to remove heavy metals from water. An illustration of a synthetic PNMBC biosorbent is represented in Fig. 1a. The adsorption results showed fast removal kinetic (10 min) with a  $Q_{\text{max}}$  of  $757.45 \text{ mg/g}$ . The reusability investigations of PNBBC illustrate that the adsorbent material could be reused for  $\text{Cu(II)}$  with capacity  $81.26 \text{ mg g}^{-1}$ ,  $70.7 \text{ mg g}^{-1}$ , and  $75.85 \text{ mg g}^{-1}$  at (1:1, 1:2, 2:1) for, respectively, after four cycles at  $15^\circ\text{C}$ . Another effective cellulose-based material for selective com of copper was investigated by Wang et al. (2022a, b) that prepared cellulose aerogel via functionalization of nanofibrillated cellulose (NFC) with brushite, and PEI was investigated with high saturation reach to  $332.66 \text{ mg/g}$ . Moreover, the composite exhibited high efficiency in dynamic industrial wastewater treatment. The shape and selectivity properties of the prepared PNFC/BRU-0.2 remained stable even after 8 runs of reusability, where the adsorption capacity after 8 cycles was  $103.15 \text{ mg g}^{-1}$  and kept 96.7% of that obtained at first one. Due to the green and economical preparation synthetic of PNFC/BRU and its promising management character, PNFC/BRU can provide a novel adsorbent for the heavy metals. Liu et al. (2022a, b, c) modified loofah with mannitol (MML) for efficient removal of  $\text{Cu(II)}$ .

An efficient adsorbent (MML) was prepared successfully, as shown in Fig. 1b. The MML composite showed a saturation capacity ( $Q_e$ ) of  $888.9 \text{ mg/g}$ . The interaction mechanism of  $\text{Cu(II)}$  ion onto MML is cleared in Fig. 1c. At a higher pH, the complexation occurs to achieve adsorption where the O-atoms in the ( $-\text{OH}$ ) group located on cellulose molecular chains have lone pairs of electrons can be shared with the  $\text{Cu(II)}$ . Also, adsorption can be occur through chelation in addition to complexation where these hydroxyl have great tendency to chelate  $\text{Cu(II)}$  ions (Liu et al. 2022a, b, c).

**Table 1** Application of cellulose-based materials for removal of inorganic pollutants

Adsorbent	Modifier	Adsorbate	Adsorption capacity (mg/g)	Adsorption percent	Adsorption parameters	Reuse	References
Nanocellulose/magnetic bentonite	PEI	Cu <sup>2+</sup>	757.45		$t = 10$ min, pH 5, $T = 15$ °C, dose = 0.01 g, $C_0 = 100$ mg/L	4 cycles 1 mol/L NaOH	Sun et al. (2022)
NFC/brushite	PEI	Cu <sup>2+</sup>	332.66	99.99	Dose = 0.3 g, pH 3.5, $C_0 = 900$ mg/L, $T = 25$ °C, $t = 120$ min	8 cycles 0.1 M EDTA-2Na 96.7%	Wang et al. (2022a, b)
Loofah	mannitol	Cu <sup>2+</sup>	888.9	97.48	$C_0 = 900$ mg/L, pH 5, $T = 25$ °C, $t = 120$ min	5 cycles 0.1 M HCl 93.82%	Liu et al. (2022a, b, c)
Cellulose	KH-792/DTPA	Cu <sup>2+</sup>	298.62	96.58	pH 6, $t = 120$ min, $T = 40$ °C	5 cycles 0.1 M HCl 82.56%	Gao et al. (2022)
CNFB/STMP	STMP	Cu <sup>2+</sup>	141.4 (PRM) 147.9 (SM)	99	$t = 120$ min, pH 6	4 cycles 0.1 M HCl 83%	Maaloul et al. (2021)
CC-DA	p-PDA/ECH	Pb <sup>2+</sup>		98.5	$t = 15$ min, dose = 2 mg/mL, $T = 25$ °C		Jawhird et al. (2021)
Carbonized cellulosic materials	sodium acetate	Pb <sup>2+</sup>	243.5		$C_0 = 250$ mg/L, dose = 0.01 g, $t = 10$ min, $T = 25$ °C	6 cycles 10 wt% EDTA 98.5%	Ai et al. (2022)
$\beta$ -CD@MRHC	$\beta$ -cyclodextrin	Pb <sup>2+</sup> BPA	266.2 412.8		$t = 30$ and 7.5 min, $T = 25$ °C, pH = 7	3 cycles 1 mol/L HCl	Liu et al. (2022a, b, c)
CNC-nZVI	nZVI	Ni		98.5%	pH 5, dose = 0.2 g/L, $C_0 = 100$ mg/L, $t = 240$ min		Song et al. (2022)
Poplar cellulose	TEMPO/PEI/GA	Cu <sup>2+</sup> Pb <sup>2+</sup>	109.89 279.32		pH 5, $t = 45$ min, $T = 25$ °C	5 cycles Cu(II) 58.26 mg g <sup>-1</sup> Pb(II) 91.96 mg g <sup>-1</sup>	Xing et al. (2021)
M-HCS/MCCs	HCS	Pb <sup>2+</sup> Cu <sup>2+</sup> MO CR	412.50 425.14 435.13 538.46		$t = 60$ and 30 min, dose = 30 mg,		Zeng et al. (2023)
MSB	1,2,3,4-butane-tetracarboxylic acid dianhydride	Cd <sup>2+</sup> Pb <sup>2+</sup>	0.50 mmol g <sup>-1</sup> 0.61 mmol g <sup>-1</sup>				Elias et al. (2022)
HEMA/C	HEMA	Cd <sup>2+</sup> Pb <sup>2+</sup>	901 926		pH 5, $t = 180$ min, Co = 30 mg/L		Gouda and Aljaafari (2021)
Cellulose acetate sponges	Cyanex 923®	Cd <sup>2+</sup> Zn <sup>2+</sup>	8.90 45.43			25 mol L <sup>-1</sup> HNO <sub>3</sub>	Páez-Hernández, et al. (2021)

Table 1 (continued)

Adsorbent	Modifier	Adsorbate	Adsorption capacity (mg/g)	Adsorption percent	Adsorption parameters	Reuse	References
Fe <sub>3</sub> O <sub>4</sub> /CNF/PEI/SHMMT	PEI/SHMMT	Pb <sup>2+</sup> Cu <sup>2+</sup> Cd <sup>2+</sup>	429.18 381.68 299.40	85.92 87.07 86.53	$T=25^{\circ}\text{C}$ , $\text{pH}=6$ , $C_0=800\text{ mg/L}$ , $\text{dose}=0.5\text{ g/L}$ , $t=300\text{ min}$ $t=60\text{ min}$	5 cycles > 85%	Yuan et al. (2023)
CCMA	Acid activated montmorillonite	Cu <sup>2+</sup> Pb <sup>2+</sup> Cd <sup>2+</sup>	181.92 170.19 163.85		$t=60\text{ min}$	5 cycles 1.0 M Na <sub>2</sub> EDTA 80% (the defaulted first adsorption capacity)	Rong et al. (2021)
CMC/GG/MTC	KMnO <sub>4</sub>	Cu <sup>2+</sup> Co <sup>2+</sup> MB	805.45 772.52 598.28		Co = 100 mg/L for Cu <sup>2+</sup> and Co <sup>2+</sup> , and 25 mg/L for MB, dosage = 0.2 g/L, $T=25^{\circ}\text{C}$ , $t=10\text{ h}$	5 cycles 1 M HCl 82%	Yang et al. (2023a, b)
C-P	Phosphate	Ni <sup>2+</sup> Cu <sup>2+</sup> Pb <sup>2+</sup> Cd <sup>2+</sup>	40–49.4				El-Zawahry et al. (2023)
MC-Tz	Schiff base heterocyclic	Pb <sup>2+</sup> Cu <sup>2+</sup> Ni <sup>2+</sup> Cd <sup>2+</sup>	453.2 485.5 473.2 455.6		$\text{pH}=6$ , $\text{dose}=80\text{ mg}$ , $t=60\text{ min}$	4 cycles 0.1 M HCl, H <sub>2</sub> SO <sub>4</sub> , and CH <sub>3</sub> COOH	Mahalakshmi et al. (2022)
Cel-dend-SO <sub>3</sub> H	Sulfonic acid-modified dendrimer	Pb <sup>2+</sup> Na <sub>2</sub> SO <sub>4</sub> RB RB50 AZ		97% 96.9% 2.1% 8.2% 5.3%			Vatanpour et al. (2022)
LCUM/D	LDH/DA	Ti(I)		97.18%			Shao et al. (2023)
MgO@CAM	MgO	Ti(I)		97.6%	$\text{pH}=3$		Shao et al. (2021)
DMAE-PS	Zwitterionic N-(3-sulfonato-1-propyl)-N,N-dimethylammonium	Ti(I)	274.7		Dose = 1.5 mg, $\text{pH}=2$ , $t=60\text{ min}$	5 cycles 0.1 M EDTA-2Na > 90%	Yang et al. (2023a, b)
G-DMC	Diethylenetriamine-pentaacetic acid	Hg <sup>2+</sup>	443.8		$\text{pH}=4$ , $t=10\text{ min}$ , $T=25^{\circ}\text{C}$	5 cycles 0.1 mol/L HCl 88.13% of its original adsorption capacity	Li et al. (2021)
Cellulose microspheres	Guanine, xanthine, hypoxanthine and adenine	Au <sup>3+</sup>	307.69–510.20		$t=4\text{ h}$ , $\text{pH}=3$ , $T=35^{\circ}\text{C}$	5 cycles 0.5 mol/L mixture of HCl and thiourea 62–70%	Zhang et al. (2023a, b, c)
PEI/RC	PEI	Cr(VI)	578		$\text{pH}=5$ , $t=150\text{ min}$ , $T=45^{\circ}\text{C}$	7 cycles 0.1 M NaOH 90%	Kim et al. (2022)

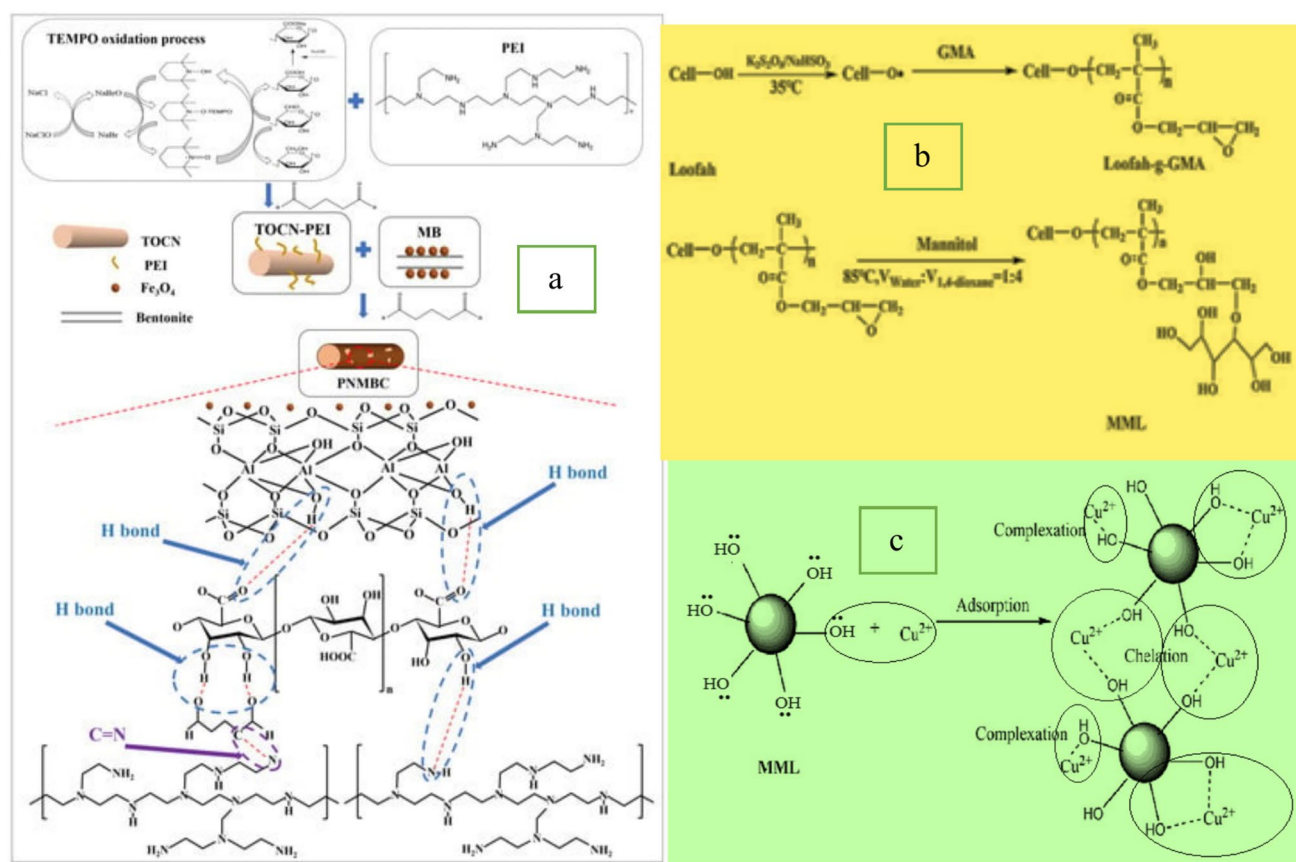
Table 1 (continued)

Adsorbent	Modifier	Adsorbate	Adsorption capacity (mg/g)	Adsorption percent	Adsorption parameters	Reuse	References
C-PEI	PEI	Cr(VI)	106.60		pH 3, $t = 180$ min, $T = 30$ °C, dose = 0.01 g,	5 cycles 0.1 M HCl 54% of its original adsorption capacity	Pattarirath et al. (2023)
cellulose modified by fibers in-depth sulfhydryl	Sulphydryl	Cr(VI)	120.60		$T = 5$ min, pH 3, dose = 50 mg; $C_0 = 50$ –500 mg/L <sup>-1</sup>		Wang et al. (2022a, b)
CA/GO/SDS	GO/SDS	P		87.2%	pH 7, dose = 0.05 g, $t = 180$ min, $C_0 = 15$ mg/L		Amin et al. (2021)
TFC/BA-CNC	BA	F <sup>-</sup>		97.0%			Huang et al. (2023a, b)
TFC/APTES-CNC	APTES						
OCNF-AgMgOnHaP	MgO/HaP	F <sup>-</sup>	8.71	99	pH 5, dose = 0.25 g, $t = 10$ min, $T = 25$ °C	4 cycles 0.01 M NaOH and 0.1 M Na <sub>2</sub> CO <sub>3</sub> $\approx 47\%$	Ayinde et al. (2022)

Facile preparation, economic, and reusable cellulose-based material were investigated by loaded cellulose with *N*-[3-(trimethoxysilyl)propyl]ethylenediamine (KH-792) followed by amidation with diethylenetriaminepentaacetic acid (DTPA) (Gao et al. 2022). The modification increased Cu(II) saturation capture, reaching 298.62 mg g<sup>-1</sup> from aqueous solutions. Indeed, the modified cellulosic material shows promise as a adsorbent for Cu(II) removal from polluted water. Also, Maaloul et al. investigated the synthesis of novel nonexpensive adsorbents for scarce water treatments to develop the efficiency and eco-friendly of agro-waste-based methods where BASB/STMP and CNFB/STMP were extracted from bleached almond shell (BASB), and cellulose nanofiber from almond shell (CNFB) crosslinked with sodium trimetaphosphate (STMP) to design BASB/STMP and CNFB/STMP adsorbent beads, respectively. The isotherm model studies showed that the BASB/STMP fitted by the Redlich–Peterson model and the CNFB/STMP by the Sips model reached the removal capacities of 141.44 mg g<sup>-1</sup> and 147.90 mg g<sup>-1</sup>, respectively. Moreover, the desorption and reusability investigations of the BASB/STMP bioadsorbent offer high separation efficiency and reusability after five cycles with the desorption percent > 80.0%. Also, previous works were investigated to prepare cellulosic-based materials for wastewater remediation. By electrospinning and surface grafting with (PMAA) (poly(methacrylic acid)), (CA) nonwoven membrane was investigated as considerable adsorbents to adsorb Cu(II), Hg(II), and Cd(II) (Tian et al. 2011). The experimental results of desorption and reuse of the membrane study cleared that the removal capacities were decreased with reusing cycles for Cu(II) and Cd(II) and steadily remained with unchanged for Hg(II).

### For Pb(II)

Pb(II) contamination in wastewaters is of great concern for environment, human, animals, and other ecosystems even at certain concentrations where its toxicity is known as lead poisoning (Jawhid et al. 2021; Ai et al. 2022). A novel functional cellulose-based biosorbent produced from stem waste biomasses for wastewater remediation was investigated (Jawhid et al. 2021). The cellulose was derived from safflower (*Carthamus tinctorius* L.) stem waste and subsequently oxidized to form cellulose dialdehyde (CC-DA) (Jawhid et al. 2021). The CC-DA was linked with *p*-phenylenediamine (*p*-PDA) by reaction with epichlorohydrin (ECH). The removal of Pb(II) reached 98.5% at pH 6 and was achieved within 15 min. Also, various works were investigated to explain the efficiency of cellulose-based materials in eliminating Pb(II) from wastewater. Ai et al. (2022) examined the elimination of Pb(II) using carbonized cellulosic materials without air and engineered with an ion exchange approach. It was shown that the  $Q_{\max}$  reached



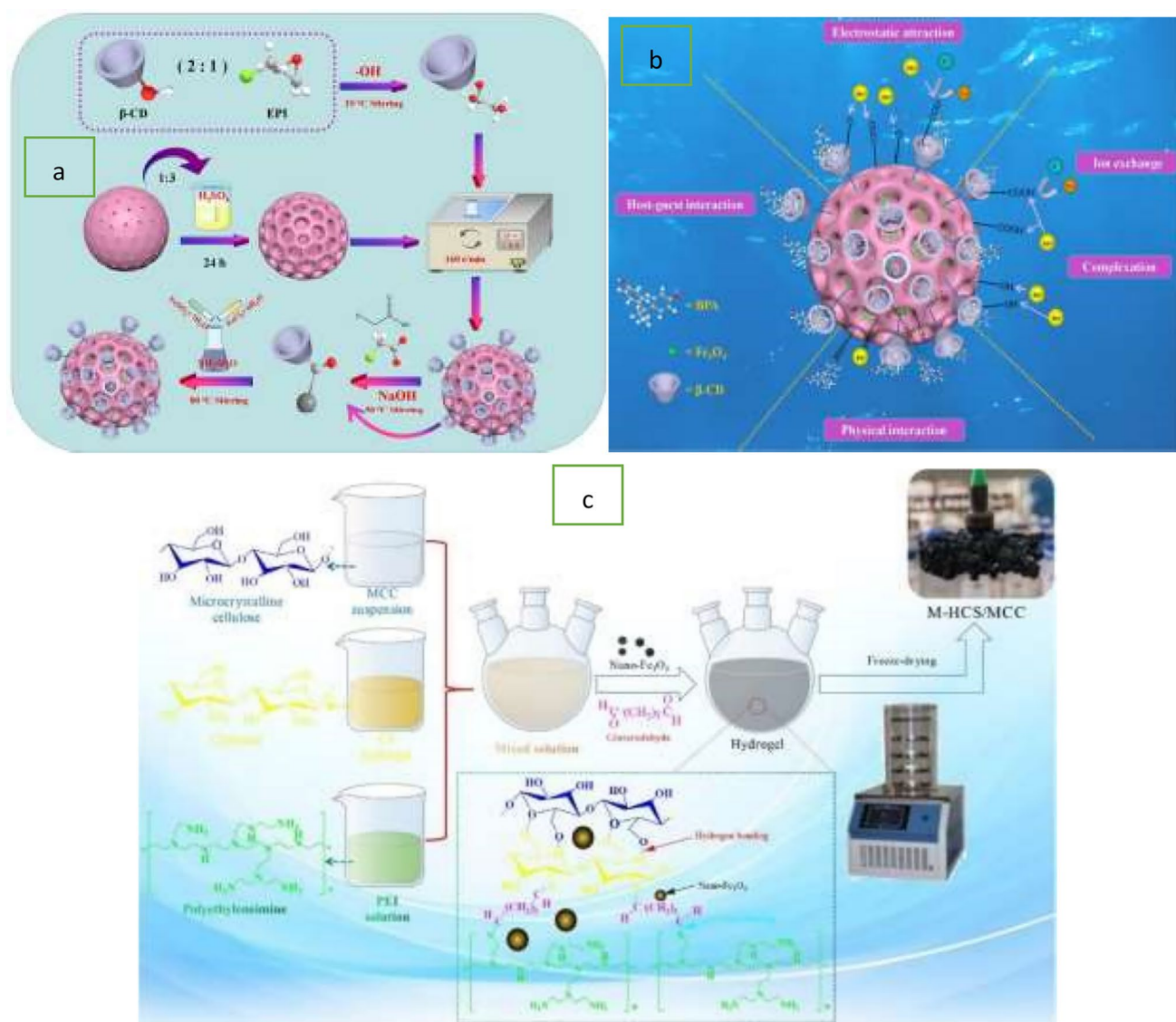
**Fig. 1** **a** Synthetic PNMBC biosorbent (this figure has been adapted/reproduced from ref. Sun et al. 2022), **b** the principle of MML preparation (this figure has been adapted/reproduced from ref. Liu et al. (2022a, b, c), and **c** mechanism of the interaction between Cu(II) ion

and MML by complexation and chelation mechanisms (this figure has been adapted/reproduced from ref. Liu et al. (2022a, b, c) all with MDPI permission, copyright 2022),

up to 243.5 mg/g through activation with sodium acetate. The regeneration study of NaAc/MCC-C material exhibited prominent regeneration and reusability after five times.

Furthermore, the adsorbed Pb(II) could be completely recovered using EDTA, and the removal rate was maintained constant after five runs of reuse. Another promising adsorbent based on cellulosic materials extracted from rice husk (RH) used for simultaneous Pb(II) adsorption from wastewater with high performance was investigated (Liu et al. 2022a, b, c). This work modified RH cellulose via anchoring with  $\beta$ -cyclodextrin ( $\beta$ -CD@MRHC) for chelation with Pb(II) and BPA. The synthesis of  $\beta$ -CD@MRHC and the proposed removal mechanism for Pb(II) and BPA are illustrated in Fig. 2a and b, respectively. The  $\beta$ -CD@MRHC exhibited uptake capacity for Pb(II)/BPA of 266.2 and 412.8 mg/g and fast elimination kinetic up to 30 and 7.5 min, respectively. In addition, after three desorption stages for loaded  $\beta$ -CD@MRHC, the desorption performance of Pb(II) and BPA reached to 76.44% and 86.22%, respectively. Further works were published to explain the preparation of a poplar cellulose-based adsorbent for the elimination of Cu(II) and Pb(II)

(Xing et al. 2021). This work investigated the activation of cellulose in the NaOH/urea/water system and regeneration in hydrogel form. After that, the cellulose hydrogel was oxidized by TEMPO and accelerated by microwave. Finally, the oxidized hydrogel (TCH) is grafted with PEI in the addition of glutaraldehyde as a crosslinking agent to yield TCP. The highest chelation capacities of Cu(II) (109.89 mg/g) and Pb(II) (279.32 mg/g) were obtained by TCP (Xing et al. 2021). Moreover, TCP's Pb(II) adsorption rate was faster than that of Cu(II). On the other hand, the adsorption capacities of Cu(II) and Pb(II) onto TCP after five cycles were 58.26 mg g<sup>-1</sup> and 91.96 mg g<sup>-1</sup>, respectively (Xing et al. 2021). Zhen et al. (2023) fabricated two novel aerogels based on microcrystalline cellulose anchored with hyperbranched chitosan (HCS/MCCs) and magnetic composite containing nano-Fe<sub>3</sub>O<sub>4</sub> particles (M-HCS/MCCs) for removal of pollutants. The two aerogels that were fabricated presented a fast and effective removal ability for dissociated pollutants (Pb(II) and Cu(II)). The schematic diagram of the microcrystalline cellulose-modified hyperbranched chitosan magnetic aerogels (M-HCS/MCCs) fabrication is illustrated



**Fig. 2** **a** Synthesis of  $\beta$ -CD@MRHC (This figure has been adapted/reproduced from ref. Liu et al. 2022a, b, c, **b** proposed removal mechanisms for Pb(II) and BPA onto  $\beta$ -CD@MRHC (This figure has been adapted/reproduced from ref. Liu et al. 2022a, b, c, both with Elsevier

permission, copyright 2022), **c** preparation of microcrystalline cellulose-modified hyperbranched chitosan magnetic aerogels (M-HCS/MCCs) (Zhen et al. 2023). (This figure has been adapted/reproduced from ref. 24, Elsevier permission, copyright 2023)

in Fig. 2c. The obtained results confirmed that in a single system, the rHCS/MCCs showed suitable adsorption rate of Pb(II), Cu(II), methyl orange (MO) and Congo red (CR), within 240 min with maximum uptake capacity of 412.50, 425.14, 435.13, and 538.46 mg/g, respectively. Furthermore, in binary contaminated environments, the M-HCS/MCCs showed superior degradation in Cu(II)-MO and Pb(II)-CR. Other cellulosic-based materials such as S-ligand tethered cellulose nanofibers (CNFs) were fabricated to adsorb Pb(II) and Cd(II) from industrial and synthetic wastewaters (Abu-Danso et al. 2018). The uptake capacities obtained were 1.16 mmol/g for Pb(II) and 0.82 mmol/g for Cd(II). The reusability studies illustrated that the prepared CNFs were

still remained have consistent elimination performance for Pb(II) and Cd(II) ions after four cycles.

### For Cd(II)

The fabrication of sustainable adsorbent materials with enhanced ability has gained scientists' attention due to the environmental contamination caused by wastewater pollution and a shortage of water resources (Elias et al. 2022; Gouda and Aljaafari 2021; Páez-Hernández et al. 2021; Yuan et al. 2023; Rong et al. 2021; Yang et al. 2023a, b; El-Zawahry et al. 2023; Adewuyi and Oderinde 2021; Mahalakshmi et al. 2022). Therefore, an urgent need is to investigate

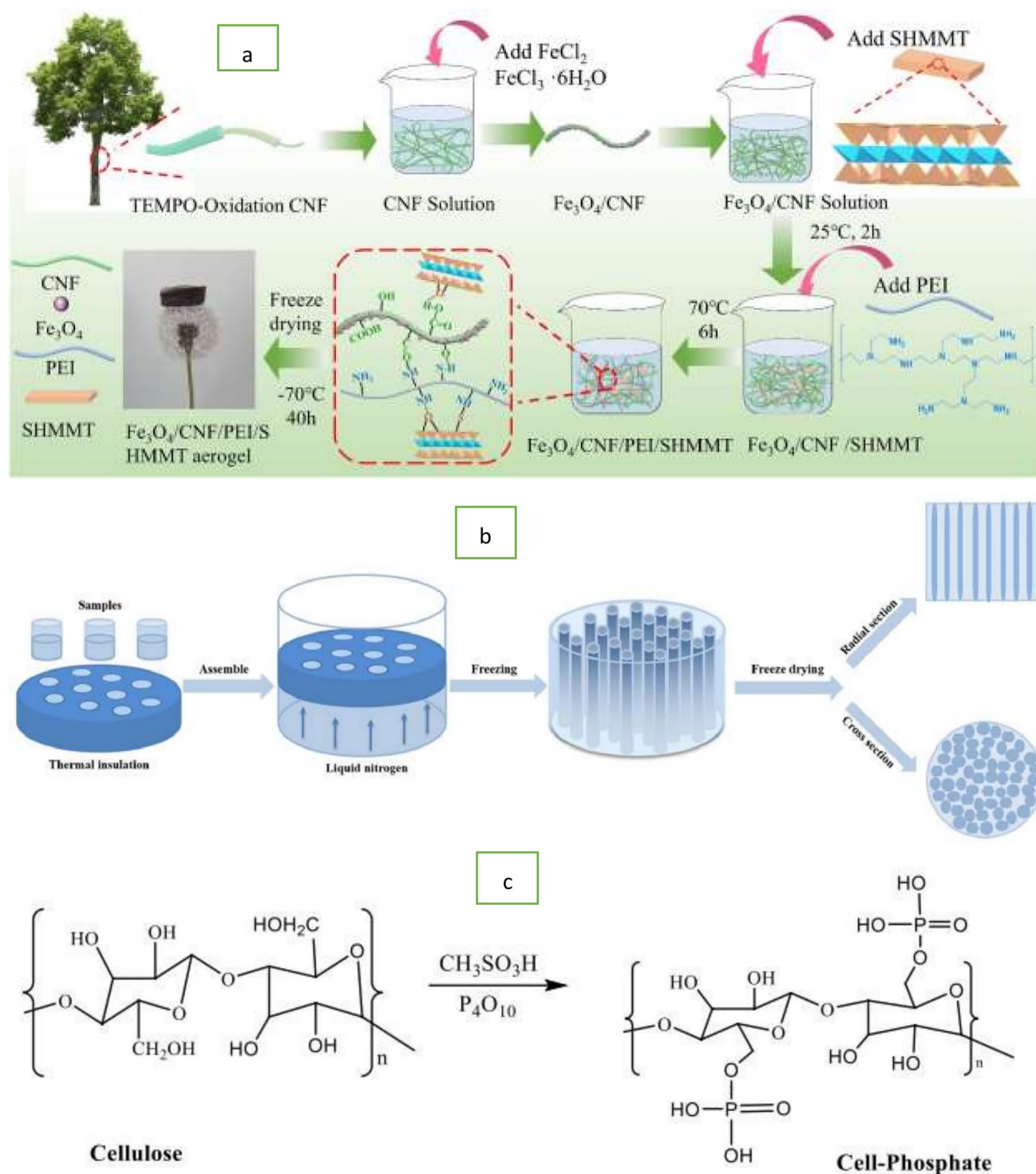
or use various biomaterials to fabricate the ideal adsorbents due to their higher toxicity Cd(II), potentially hazardous if discharged in large quantities into the surrounding environment. Therefore, there is a market demand to develop eco-friendly and lower-cost technologies to purify wastewater from such contaminants. Recently, the synthesis of a new sugarcane bagasse (SB) modified with 1,2,3,4-butane-tetracarboxylic acid dianhydride to form (MSB) for capture of Cd(II) and Pb(II) from wastewater was studied (Elias et al. 2022). The results obtained illustrated that the reusing and regeneration of prepared MSB-adsorbent material in different adsorption cycles can be regarded as advantage for the proposed technology. The fabricated cellulose-based materials typically used as biosorbents could enhance wastewater treatment technology's economic feasibility. Also, the modified cellulose nanofibers (PHEMA/C) grafted with 2-hydroxyethyl methacrylate (HEMA) through free radical copolymerization initiated by ceric ammonium nitrate (CAN) under microwave radiation were investigated for the elimination of Cd(II) and Pb(II) from wastewater (Gouda and Aljaafari 2021).

The fabricated electrospun PHEMA/C nanofibers showed effectiveness in the uptake of Cd(II) and Pb(II) at high and low environmental concentrations (Gouda and Aljaafari 2021). Modified cellulose acetate sponges derived from cigarette butts loaded with Cyanex 923 were synthesized to recover Cd(II) and Zn(II) from aqueous media (Páez-Hernández et al. 2021). For preformed uptake of Cd(II), Cu(II), and Pb(II) from aqueous media, the magnetic hybridized aerogel  $\text{Fe}_3\text{O}_4/\text{CNF}/\text{PEI}/\text{SHMMT}$  consisted of  $\text{Fe}_3\text{O}_4$  particles, cellulose nanofiber, PEI, and thiol-modified montmorillonite was prepared (Yuan et al. 2023), as illustrated in Fig. 3a. The aerogel possessed high porosity with effective chelating sites represents the smart solution in facilitated removal of the metal ion with large capacities reach to 429.18, 381.68, and 299.40 mg/g for Pb(II), Cu(II), and Cd(II), respectively. In addition, incorporating  $\text{Fe}_3\text{O}_4$  improved the mechanical properties of aerogel hybrid material. After 5 adsorption/desorption stages/cycles, the removal rate of the aerogel is > 85% (Yuan et al. 2023). Another novel, eco-friendly, and simple liquid nitrogen directional freezing process was proposed to synthesize cellulose nanofiber/chitosan/montmorillonite aerogel (CCMA) through the addition of the acid-activated montmorillonite to cellulose nanofiber/chitosan system followed by freezing in liquid nitrogen to uptake Pb(II), Cu(II), and Cd(II) (Rong et al. 2021). Scheme for the fabricated porous cellulose CCMA aerogel is represented in Fig. 3b. The characterization results showed that CCMA exhibited a homogeneous 3D pore structure, high stability in harsh acid, and can withstand heavy objects 1124 times its weight (Rong et al. 2021). Further, the CCMA achieved good capture performance for Cu(II), Pb(II), and Cd(II), and reusability was maintained at 80% after 5 cycles (Rong

et al. 2021). Also, modified cellulose extracted from the seed of *Polythia longifolia* bearing phosphate functional groups to yield cell phosphate (CP), as illustrated in Fig. 3c, was investigated as an excellent agent for purification of wastewater from Cd(II), Ni(II), Cu(II), and Pb(II), respectively. For the adsorption/desorption study of Cell-Phosphate, the desorption capacities after the first cycle were in arrangement Ni(II) (90%) > Cu(II)(85%) > Cd(II)(73.05%) > Pb(II) (65.11%) (Adewuyi and Oderinde 2021). Another modified cellulose with Schiff base, such as heterocyclic interacting groups (MC-Tz), was fabricated as a non-toxic and environmentally friendly biodegradable adsorbent for Cd(II), Ni(II), Cu(II), and Pb(II) removal from wastewater. The studies of regeneration and desorption over 4-cycles demonstrated the feasibility of the sorbent's regeneration potential (Mahalakshmi et al. 2022). A modifier (charged and hydrophilic) of cellulose acetate membranes was fabricated by grafting cellulose acetate membranes with sulfonic acid-modified dendrimer (Cel-dend- $\text{SO}_3\text{H}$ ) (Vatanpour et al. 2022). Significant separation efficiency and filtration capacity were obtained through improved hydrophilic properties. Numerous doses of Cel-dend- $\text{SO}_3\text{H}$  (0.10, 0.25, 0.50, 1.0, and 2.0 wt. %) were tested for membrane preparation via the phase inversion method. The results indicated that the Cel-dend- $\text{SO}_3\text{H}$  (0.10 wt. %) possessed high water flux (82.7 L/m<sup>2</sup> h) compared with the native cellulose acetate membrane—moreover, the 2.0 wt. % Cel-dend- $\text{SO}_3\text{H}$  membrane exhibited the highest capture activity against different pollutants. In addition, all membranes showed higher sweeping of Pb(II) at 97%, with a high  $\text{Na}_2\text{SO}_4$  retention of 96.9% compared to the original membrane. Additionally, the capture of rose bengal, reactive blue 50, and azithromycin improved by 2.1%, 8.2%, and 5.3%, respectively, compared to the bare membrane, and flux and porosity were enhanced. Moreover, 2.0 wt. % Cel-dend- $\text{SO}_3\text{H}$  membrane showed an improved flux recovery ratio (FRR) raised to 92.4% compared to the value for the original membrane (73.9%).

## For Tl

Thallium has seriously impacted humans due to the increasingly serious pollution in aqueous environments. Thallium (Tl) possesses higher toxicity (> 103) than heavy metals such as Pb, Cd, Cr, Sb, Hg, and As, respectively. Tl is used in various applications and generated in wastewater streams. So, strict emission guidelines were imposed in many countries to control full (Tl) pollution, which is significant worldwide (Shao et al. 2023, 2021; Yang et al. 2023a, b). So, removing Tl from wastewater is important for environmental protection and green development (Shao et al. 2021). However, the literature showed restricted information about uptake studies compared to conventional heavy metal ions (Yang et al. 2023a, b). Recently, various types of eco-friendly materials

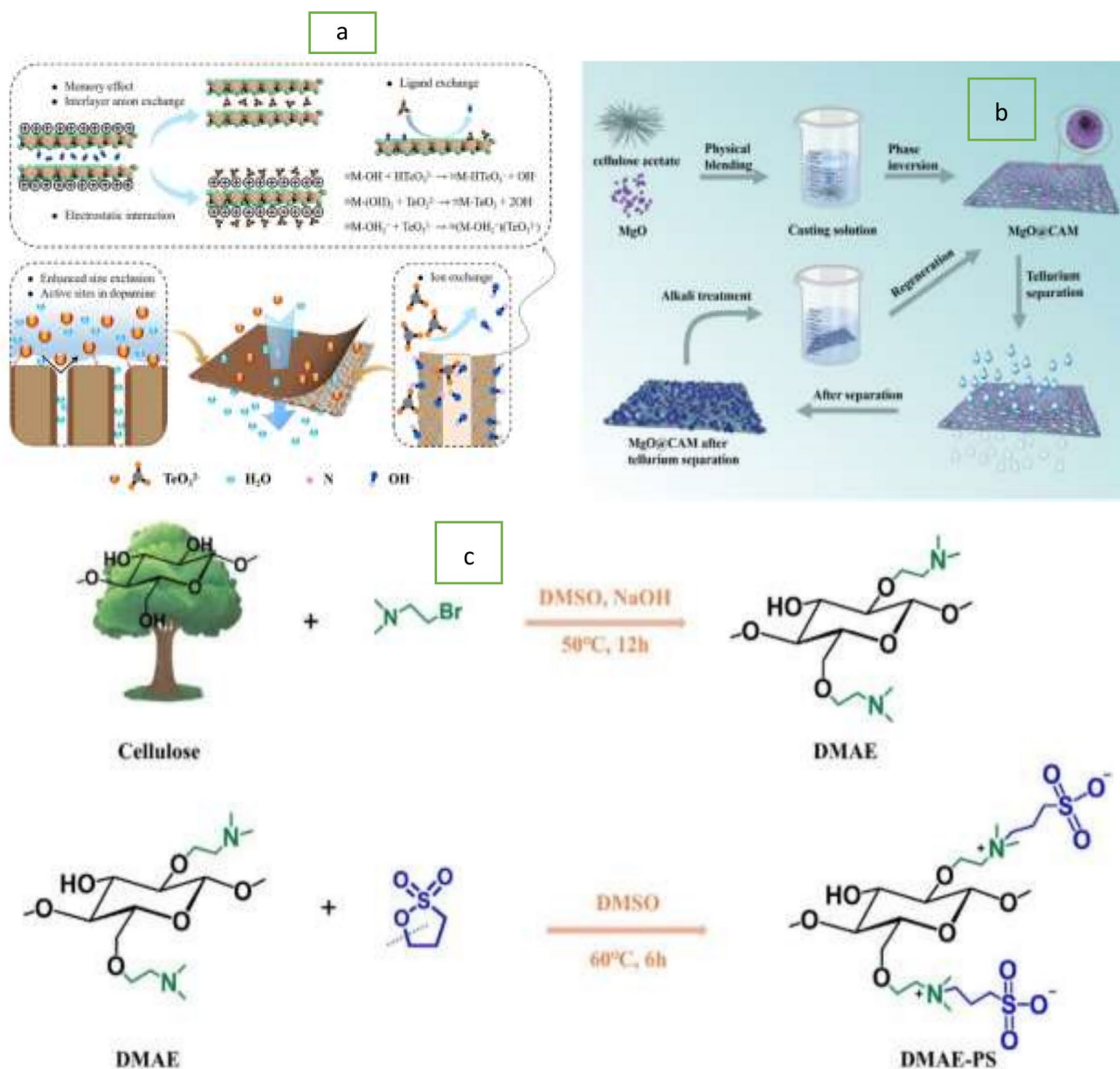


**Fig. 3** **a** Preparation process of  $\text{Fe}_3\text{O}_4/\text{CNF}/\text{PEI}/\text{SHMMT}$  (This figure has been adapted/reproduced from ref. Yuan et al. 2023, Elsevier permission, copyright 2023), **b** directional pores formation process (This figure has been adapted/reproduced from ref. Rong et al. 2021, Else-

vier permission, copyright 2021), **c** synthesis of cell phosphate (This figure has been adapted/reproduced from ref. Adewuyi and Oderinde 2021 with permission from Springer Nature Switzerland AG 2021, copyright 2021)

were fabricated to enhance the elimination of TI from aqueous solutions. A continuous 2D membrane (LCUM/D) based on (CNFs) as the support network for immobilizing layered

double hydroxides (LDHs) and dopamine (DA) as a linker was synthesized for purification of wastewater from TI, as presented in Fig. 4a (Shao et al. 2023). The results cleared



**Fig. 4** **a** Mechanism of LCUM/D for tellurium removal (This figure has adapted/reproduced from ref. Shao et al. 2023 with permission from Elsevier, copyright 2023), **b** preparation of MgO@CAM and process of tellurium separation (This figure has been adapted/

reproduced from ref. Shao et al. 2021 with permission from Springer Nature, copyright 2021), and **c** synthetic of DMAE-PS (This figure has been adapted/reproduced from ref. Yang et al. 2023a, b with permission from Elsevier, copyright 2023)

uniform coverage of the CNF skeleton with LDHs, and the defects were repaired with DA. Simultaneously, the dynamic LCUM/D membrane exhibited significant separation efficiency (97.18%) and selectivity to Tl(I). Furthermore, the LCUM/D membrane showed high stability, improved hydrophilicity, and appropriate antifouling properties, facilitating long-term applications. Another fabricated porous cellulose acetate membrane-anchored with magnesium oxide (MgO@CAM) was produced by mixing and using a phase inversion approach and investigated for tellurium recovery. Due

to the rich H-bonding of CAM, it showed good compatibility and thermal stability (Shao et al. 2021). Figure 4b points the fabrication of the MgO@CAM membrane and Tl separation. Moreover, the interconnected porous structure with a high population of OH groups makes the MgO@CAM surface highly hydrophilic with an excellent antifouling property exhibiting high removal efficiency of Tl(I), reaching 97.6% (Shao et al. 2023). Other prepared cellulose fibers grafted with zwitterionic N-(3-sulfonato-1-propyl)-N,N-dimethylammonium (DMAE-PS) were fabricated for

effective Tl sorption, as shown in Fig. 4c (Yang et al. 2023a, b). The removal was fitted with the Freundlich model with a 274.7 mg (Tl(I)) g<sup>-1</sup> saturation ability for DMAE-PS. Moreover, the composite showed suitable selectivity toward Tl(I) in the existence of metals such as Zn<sup>2+</sup>, Cr<sup>3+</sup>, Mn<sup>2+</sup>, Cu<sup>2+</sup>, and Cd<sup>2+</sup>, respectively. After five adsorption–desorption cycles, over 90% of Tl(I) was regained and the regenerated DMAE-PS after each cycle remained has high sorption capacities (225–248 mg g<sup>-1</sup>). Also, various cellulose-based materials were fabricated to treat wastewater from other inorganic pollutants (Li et al. 2021; Zhang et al. 2023a, b, c; Kim et al. 2022; Pattarath et al. 2023; Wang et al. 2022a, b; Nakakubo et al. 2022; Amin et al. 2021; Huang et al. 2023a, b; Ayinde et al. 2022).

Modification of cellulose with the diethylenetriamine-pentaacetic acid using two approaches, including catalyzed-grafting G-DMC and coupling by epichlorohydrin C-DMC, was investigated to improve the adsorption efficiency of Hg(II) from aqueous environment (Li et al. 2021). Cellulose microspheres were anchored with four nucleobases: guanine, xanthine, hypoxanthine, and adenine via radiation approach to recover Au (III) from the actual gold slag solution (Zhang et al. 2023a, b, c).

Regenerated nanocellulose hydrogel modified with cationic binding sites (Kim et al. 2022), cellulose anchored with PEI using microwave methodology (Pattarath et al. 2023), and in-depth sulfhydryl-loaded cellulose fibers (Wang et al. 2022a, b) were synthesized and used to uptake of Cr (VI) from aqueous solution. Crosslinked dithiocarbamate-modified cellulose was prepared as a sorbent of arsenate As(III) ions (Nakakubo et al. 2022). Cellulose acetate/graphene oxide/sodium dodecyl sulfate (CA/GO/SDS) was fabricated using an electrospinning approach to recover phosphate (Amin et al. 2021).

### For F(-1)

The preparation of a three-layer film (TFC) composite forward osmosis (FO) membrane was investigated for the rejection of fluoride ions (Huang et al. 2023a, b). First, the porous substrate was loaded with interlayer CNC to improve its hydrophilicity and rejection activity (TFC/CNC). After that, chemical functionalization using boric acid (BA) (TFC/BA-CNC) and (3aminopropyl)triethoxysilane (APTES) (TFC/APTES-CNC) was carried out. Modifying the membrane with CNC interlayer improved hydrophilicity and increased the surface negativity, leading to the enhanced flux of H<sub>2</sub>O molecules and the rejection of F<sup>-</sup> ions (Huang et al. 2023a, b). Biosynthesized nanofibrous cellulose template that incorporated the Ag–MgO–nanohydroxyapatite (CNF-AgMgOnHaP) was investigated as a multifunction adsorbent for hydrothermal bioreduction method for effective fluoride adsorption (Ayinde et al. 2022). The sorption of F<sup>-</sup> onto the

CNF-AgMgOnHaP was well ascribed by the Freundlich isotherm model with a maximum loading of 8.71 mg/g. Moreover, the prepared materials possessed bacterial resistance toward common infectious microbes that polluted drinking water. The reusability and regeneration study of CNF-AgMgOnHaP demonstrated that the prepared material has a greater economic potential to remove and recover of F(-1) and its reusability was reduced with an increasing regeneration cycle (Ayinde et al. 2022).

## Cellulose-based materials for oil/water separation

Oil effluent typically occurs during production, such as pipeline leaks, loading and unloading, offshore oil drilling, storage, refining, and transportation. Also, dispersal, solidification, skimming, and in situ combustion have been used to treat marine oil spills. The presence of oil films on the water level can cause damage to the aquatic systems by lowering the amount of dissolved oxygen and retard the rehydration process. Hence, increasing studies were directed to eliminate and control such pollution. During the last decades, researchers have regarded the effective oil/water mixtures separation of as a major challenge, and therefore, various solutions to these issues have been researched using different technologies (Muharja et al. 2023; Phat et al. 2021; Liu et al. 2022a, b, c; Feng et al. 2022). Therefore, it is desirable to investigate and discover affordable and eco-friendly materials that are highly efficient for oil/water separation systems with biodegradation characteristics, good environmental compatibility, low cost, and high effectively. Table 2 represents the recent utilization of cellulose-based materials for separating the oil phase from contaminated water (Muharja et al. 2023; Liu et al. 2022a, b, c; Feng et al. 2022; Chen et al. 2023; Lang et al. 2023; Panda and Gangawane 2023; Li et al. 2023; Lei et al. 2023; Huang et al. 2023a, b; Wahid et al. 2022; Francis et al. 2023; Ning et al. 2023; Sultana and Rahman 2022; Zhang et al. 2021a, b; Nabiev et al. 2023; Xi, et al. 2021; Alazab and Saleh 2022; Chu et al. 2022; Wang et al. 2021b, a).

### Cellulose-based materials aerogel

Cellulose aerogel materials are extensively used in research fields such as oil/water separation (Muharja et al. 2023; Phat et al. 2021). Recently, aerogels are widely utilized to separate oils from oil/water systems due to their great porous structure, surface area, and low density (Feng et al. 2022). On the other hand, bio-based aerogels, such as cellulose, which is most available and highly distributed in nature, have gained great attention and become more attractive in both of environmental sciences and industrial productions

**Table 2** Application of cellulose-based materials for separation of oil phase from wastewater

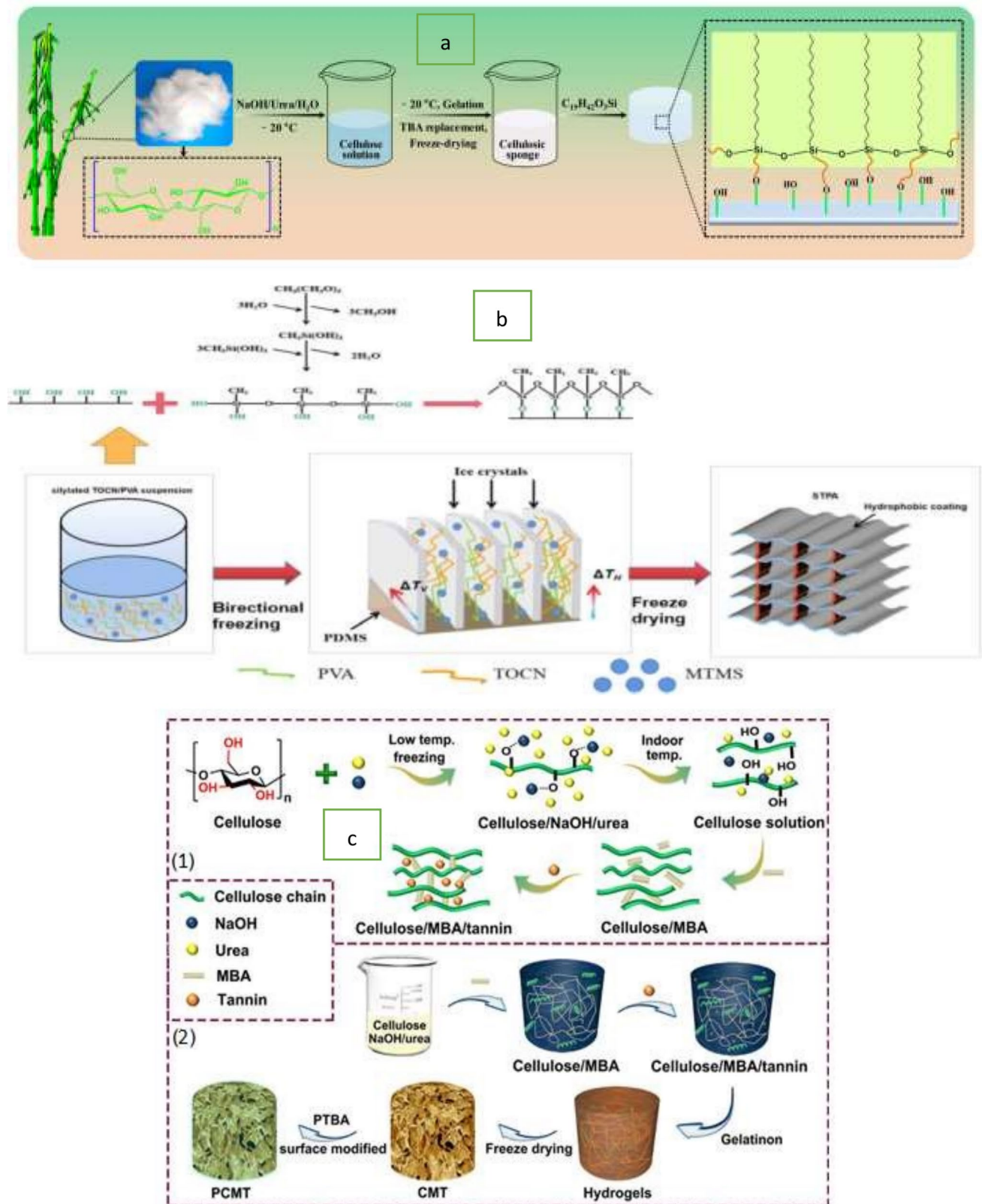
Adsorbent	Modifier	Adsorbate	Adsorption capacity (mg/g)	Separation percent	References
Aerogel composed of ramie pulp	MBA	Oil	5.81		Muharja et al. (2023)
Bamboo cellulose foam		Oil	11.5~37.5		Liu et al. (2022a, b, c)
STPA	Methyltrimethoxysilane	Different kinds of organic solvents	51–111		Feng et al. (2022)
HMCL-aerogel	(NDM) and Fe <sub>3</sub> O <sub>4</sub> NPs	O/W		99%	Chen et al. (2023)
PCMT	Tertbutyl acrylate				Lang et al. (2023)
HRCSS	Methyltrimethoxysilane	Oil	31.67–48.25	94%	Panda and Gangawane (2023)
SA/CMC	Methyltrimethoxysilane	Silicone oil	51.22		Li et al. (2023)
Cellulose cryogel	Methyltrimethoxysilane	Organic solvent and oils		90	Lei et al. (2023)
BCAs	Methyltrimethoxysilane	O/W			Huang et al. (2023a, b)
BC	TiO <sub>2</sub> /ZnO	Different oil (such as toluene, n-decane, and hexane) in water		> 99.9%	Wahid et al. (2022)
GO-CA	GO	n-hexane/water emulsion		3820 LMH	Francis et al. (2023)
d-CA/BC@CCA	BC/CA	O/W		98.34%	Ning et al. (2023)
CA/zeolite	Zeolite	Lubricating oil emulsion and vegetable oil		97%	Sultana and Rahman (2022)
(ACNTs) APTES/MXene	APTES/MXene	Oil–water emulsion		99%	Zhang et al. (2021a, b)
e-ACNTs supported by CA					
Treated cellulose acetate membranes					
Paper sheets	1,2,3,4-butanetetracarboxylic acid				
MCF	Cellulose-decafluorobiphenyl			99.3	Xi et al. (2021)
				> 97.68%	Alazab and Saleh (2022)
PDMS-Ni-PW	PDMS-Ni	Crude oil/water mixture	99.92%		Chu et al. (2022)
(CS)-(CNCs)-(SSM)	CS/CNCs	oil-in-water emulsions	99.58%		Wang et al. (2021b, a)

due to their eco-friendly environmental friendliness, recyclability, renewability, and abundance (Chen et al. 2023; Lang et al. 2023).

One of the current works published recently is focused on studying the enhanced oil removal from water by using fabricated aerogel composed of ramie pulp dissolution in NaOH-urea,  $\text{Fe}_3\text{O}_4$ , potassium peroxydisulfate, GO, and *N,N'*-methylene bisacrylamide (MBA at 1–13 g/L concentrations) (Muharja et al. 2023). The prepared magnetite cellulose aerogel/MBA/GO was bolstered aerogel structural, rendering it to reuse for 2-cycles and to offer a biodegradable cellulosic-based materials to oil spill challenges.

Cellulose aerogel (Cell-A) is an excellent method to fabricate adsorbent materials for oil recovery from oil/water systems (Phat et al. 2021). Cell-A was first prepared from Vietnamese water hyacinth, and the PVA was employed as a crosslinker using the freeze-drying method. Consequently, the Cell-A was dip-coated with poly(dimethylsiloxane) (PDMS) and calcined to form PDMS-coated cellulose aerogels (Cell-AP) and carbon aerogels (CA), respectively. Using the cellulose's 15: 1 mass ratio to PVA, CA showed higher oil adsorption capacity than Cell-AP (Phat et al. 2021). On the other hand, many works applied various sol–gel, freeze-drying, and surface hydrophobic functionalization approaches for the synthesis of a foam like structure of bamboo cellulose with superior wetting properties (Liu et al. 2022a, b, c); the superhydrophobic/superoleophilic bamboo cellulose sponge is represented in Fig. 5a. As a result, the foam showed the WCA at  $160^\circ$ , and its superoleophilic ability promoted oil absorption with a capacity reaching 11.5–37.5 g/g for different oil types. Moreover, the foam demonstrated an excellent recycling performance reached to 10 cycles, with an oil (1,2-dichloroethane) separation capacity of up to 31.5 g/g after 10 cycles (Liu et al. 2022a, b, c). The fabrication process of silylated TEMPO-oxidized cellulose nanofibers/PVA aerogels (STPA) uses a three-step method including aqueous silylation, bidirectional freezing, and freeze-drying, respectively (Feng et al. 2022). The bidirectional freezing approach was applied to yield superior aerogels structurally by controlling the methyltrimethoxysilane (MTMS) concentration. Figure 5b illustrates the mechanism scheme of STPA (Feng et al. 2022). The STPA displayed favorable features, such as low density ( $9.79 \text{ mg/cm}^3$ ), hydrophobicity ( $136^\circ$ ), compressibility, and high adsorption capacity (51–111 g/g) for different kinds of organic solvents. Furthermore, the excellent compressibility of STPA provided a simple mechanical squeeze of absorbed oil during the recovery process. Moreover, it displayed a high oil recovery rate (80%) and is still at (90%) after 50 cycles of absorption–squeezing (Feng et al. 2022). The fabrication of hydroxyethyl cellulose–lignin aerogel (CL-aerogel) through ultrasound to develop the capability of oil/water separation was also investigated (Chen et al. 2023).

This study fabricated CL-aerogel by mixing hydroxyethyl cellulose and lignin applying a sol–gel approach followed by freeze-drying. Adding lignin provides the 3D porous structure of the CL-aerogel and improved mechanical characteristics (Chen et al. 2023). In addition, by applying ultrasound, the CL-aerogel is functionalized with *n*-dodecyl mercaptan (NDM) and  $\text{Fe}_3\text{O}_4$  NPs to produce the HMCL-aerogel with enhanced hydrophobicity and magnetic properties. As a result, the HMCL-aerogel possessed preformed oil absorption capacity with high selectivity. Meanwhile, the aerogel showed high separation efficiency of oil/water with a high flux of  $2986 \text{ L m}^{-2}\text{h}^{-1}$ , even in corrosive conditions. Furthermore, The HMCL-aerogel could be recovered by applying an external magnetic field and recycled through an easy extrusion treatment, resulting in a separation percent steady over 99% after 10 cycles (Chen et al. 2023). Utilization of sol–gel strategy for preparation of 3D porous cellulose/*N,N'*-methylenebisacrylamide/tannin (PCMT) composite modified with tertbutyl acrylate was investigated for the removal of marine or industrial oil, as shown in Fig. 5c, (Lang et al. 2023). PCMT showed significant oil–water separation ability and performed compressive strength. The high porosity of the PCMT enhanced the adsorption abilities of organic solvents, as shown in Fig. 5c (Lang et al. 2023). The PCMT porous materials showed still high hydrophobicity and oil absorption affinity under environments of high stirring, a wide pH range (1–14), a wide temperature range (4– $160^\circ\text{C}$ ), ultraviolet irradiation (8 h), and tape peeling (10 times). The reusability of the prepared material was investigated and demonstrate underwent changes across 10 cycles. It was noted that the oil absorption ability was decreased after the second cycle and remained nearly constant or slightly decreased till the tenth cycle. Panda and Gangawane (2023) were applied to freeze-dry for preparation of combined reused cellulose-silica (HRCSSs) aerogels utilizing the 3D cellular network, crosslinker (Kymene), and filler ((MTMS)-derived silica aerogels). The produced HRCSSs resulted in a stable superhydrophobic material [water contact angle (WCA) of  $163.40 \pm 2.5$ ,  $160.0 \pm 1.2$ ,  $168.0 \pm 1.5$ ] due to the silylation treatment for 1, 2, and 4 wt % loading of cellulose in combined foam. Utilizing the aerogel enhanced the porosity, superelasticity, and compressibility, while silica nanoparticles improved the oleophilic properties. The HRCSS aerogel exhibited an oil adsorption affinity within the 31.67–48.25 g/g range with 94% retention capacity, and it was reused up to 10 cycles for various wt. % of cellulose fiber concentration. Li et al. (2023) fabricated superhydrophobic sodium alginate/sodium carboxymethyl cellulose (SA/CMC) aerogel employing silica nanoparticles and (MTMS). The obtained 3D aerogel had a density of  $0.080 \text{ g/cm}^3$ , excellent porosity of 94%, water stability, compressibility, and great adsorption ability toward organic solvents (methylene blue and chloroform). The data showed that the



**Fig. 5** **a** Superhydrophobic/superhydrophilic bamboo cellulose foam (This figure has been adapted/reproduced from ref. Liu et al. 2022a, b, c with MDPI permission, copyright 2022). **b** Mechanism scheme of STPA (This figure has been adapted/reproduced from ref.

Feng et al. 2022, Springer Nature permission, copyright 2022). **c** (1) the hydrogels forming process; (2) the experimental process of the PCMT (This figure has been adapted/reproduced from ref. Lang et al. 2023, Springer Nature permission, copyright 2023)

superhydrophobic SA/CMC aerogel possessed a suitable compressive and improved adsorption capacity. Lei et al. (2023) extracted cellulose from the herbal waste (*Ficus microcarpa* L. f) for the preparation of cryogel, which was anchored with (MTMS)<sub>10</sub> to improve its hydrophobicity and minimize its surface energy. The modified cryogel showed the maximum loading capacity of silicone oil (51.22 g/g). Moreover, it can separate emulsion water in toluene stabilized by Span 80, with efficiency of 98.57% and a flux of 1474.67 L/m<sup>2</sup>h. By drying absorbed samples or mechanically squeezing out oils, the prepared aerogel could be regenerated after adsorbing organic liquids.

Although all of these works illustrate the fact that the modification step of surface is an intrinsic part of preparing the cellulose aerogel materials, there are many challenges facing the long-term stability and application potential of such biopolymers aerogel counterparts such as their intrinsic hydrophilicity (Zhao et al. 2018). Therefore, many of the commercially successful cellulose-based materials aerogels insulation product will require further modifications to resolve these limitations. Thus, long-term stabilities of cellulose-based materials aerogel require more investigations and studying by the scientific community. On the other hand, due to the organic nature of most cellulose-based materials aerogels they probably have reduced fire safety ratings compared to other aerogel materials attributing to their limited operating temperatures (Zhao et al. 2018, 2017). Therefore, the smooth transition of the further functionalization of nanocellulose aerogels from a laboratory to industrialization encountered other challenges, such as modification of functional groups, complex and time-consuming fabrication, designable structures, and undesirable dispersion of fillers, which remain unresolved. To overcome these challenges, several efforts are made to seek novel processes to improve the existing approaches (Chen et al. 2021).

### Bacterial cellulose (BC)

BC is a high molecular polysaccharide formed by some bacterial strains via fermentation (Wahid et al. 2022). During the last decades, bacterial nanocellulosic (BNC) materials have attracted more attention due to their long aspect ratio, crystallinity, and high purity (Wang et al. 2023a, b). BC aerogels were fabricated from kombucha fermentation fruit waste and freeze-dried to form 3D of (BC) for oil/water separation and thermal insulation (Huang et al. 2023a, b), as represented in Fig. 6a. The obtained aerogel possessed significantly low density (5.693 mg/cm<sup>3</sup>), enhanced porous structure (98–99%), and low thermal conductivity (23.4 mW/(m K)). After that, the BCAs modification with MTMS via chemical vapor deposition was used to optimize their organic solvent and oil adsorption. This improved compression resilience by 90% and the water contact angle to

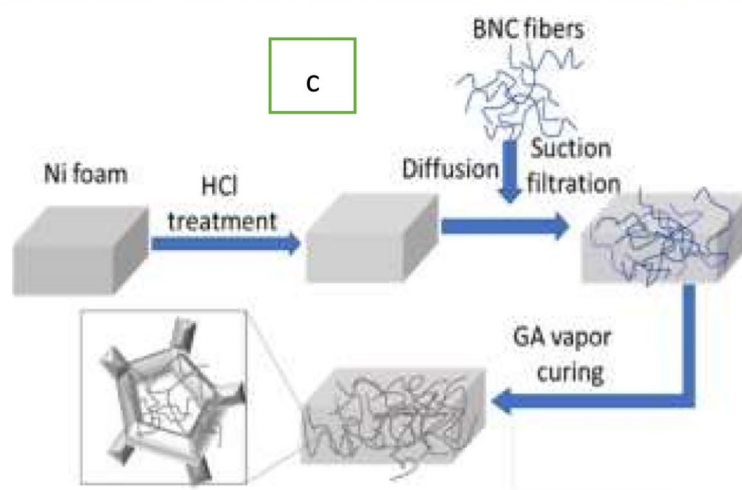
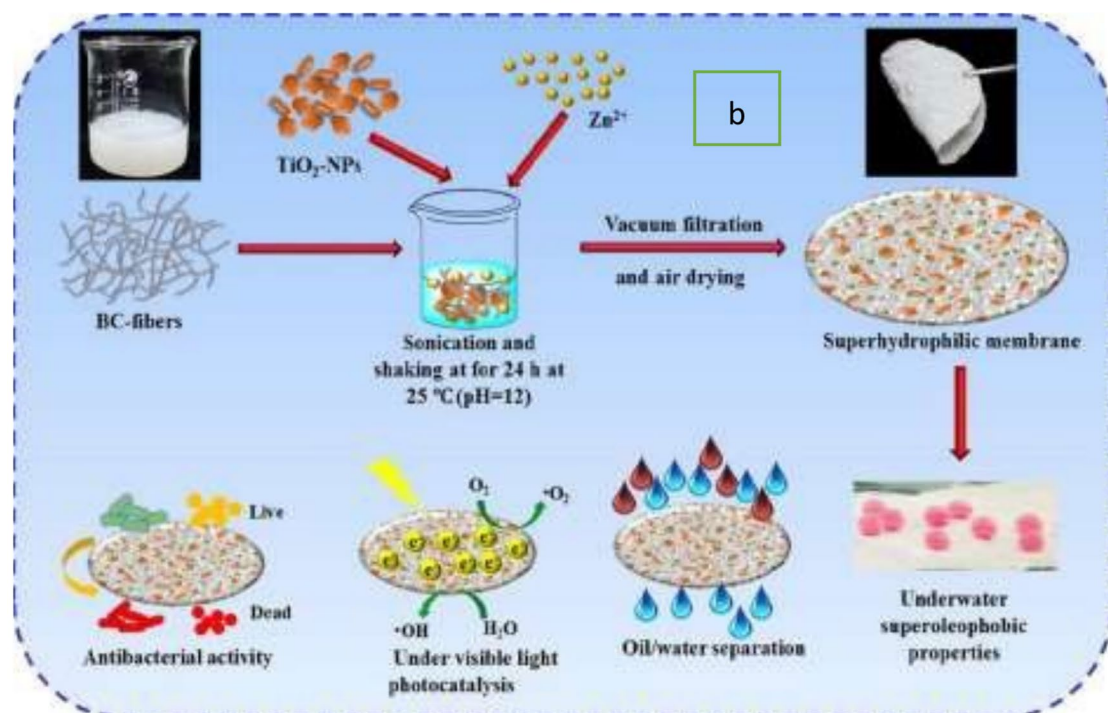
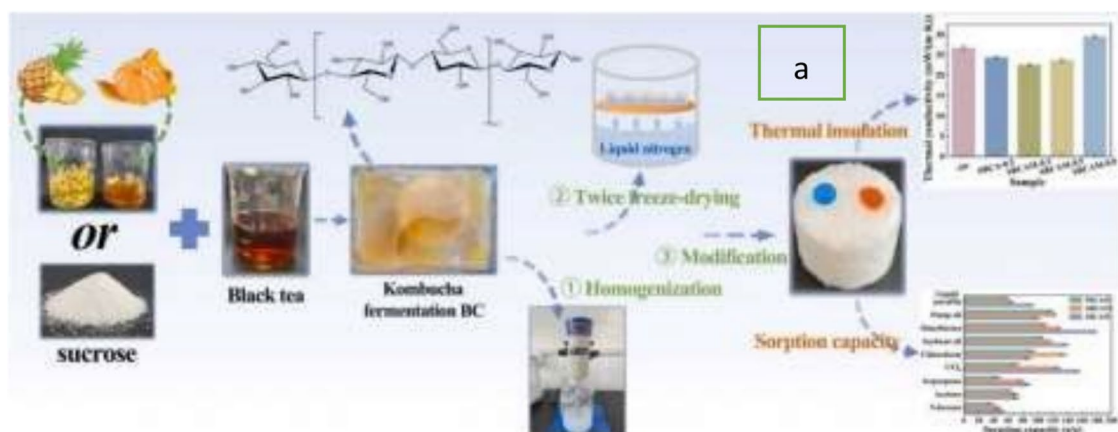
146.4° (Huang et al. 2023a, b). The reusability study of the prepared aerogel exhibited decreasing in adsorption capacity to 93.4% after six times and remained > 80 gg<sup>-1</sup> for ten cycles.

Another recent work reported the fabrication of the BC with TiO<sub>2</sub>–ZnO as a multifunctional membrane for oil/water separation by blending with BC nanofibers in the presence of TiO<sub>2</sub> and subsequently anchored with in situ ZnO–NPs growth (Wahid et al. 2022), as illustrated in Fig. 6b. Under low driving pressure (0.2–0.3 bar), the flux rate was 8232.81 ± 212 L m<sup>-2</sup> h<sup>-1</sup> and the separation performance higher than 99.9%. Furthermore, mates also could treat oil-in-water emulsion with a separation rate of 1498 ± 74 L m<sup>-2</sup> h<sup>-1</sup> and efficiency (99.25%). Moreover, the blended membrane possessed high photocatalytic efficiency (> 92%) and high antibacterial efficiency against various bacterial strains. The experimental data displayed that the reuse of the fabricated membrane was recycled 20 times with no changes observed for all cycles in the separation efficiency or flux rate. Wang et al. (2023a, b) modified porous metal foam (Ni foam plate and slice) with bacterial nanocellulose fiber (BNC) to improve its hydrophilicity.

The fiber content controlled the foam pore size, as shown in Fig. 6c. The presence of BNC reduces the large pores originating from Ni foam while keeping a high porosity level. Moreover, the thin slice Ni foam with different pore sizes highly rejected oil from various oil/water blends (i.e., layered oil/water mixtures, oil/water emulsions with and without surfactant) under gravity with a higher flux rate than thick Ni foam. The anchored Ni foams show valid regeneration after washing cycles with ethanol. At the same time, the loaded Ni foams could be used under severe water impact and large pressure differences owing to their high compressive modulus.

### Electrospinning cellulose-based materials

Electrospun functionalized membrane is a new class of versatile membrane contactors applied recently to separate the free-floating oils on oil-polluted water (Francis et al. 2023; Sultana et al. 2022). Electrospun displayed membranes have various advantages, such as improved performance of organic/inorganic layers, large surface/volume ratios, and mechanical properties (Sultana et al. 2023). Francis et al. (2023) used an electrospinning approach for the production of cellulose acetate (CA) followed by electrohydrodynamic loading of (GO) to improve its hydrophilicity that required for oil/water separation. The result showed that the WCA of the GO-CA membrane decreased from 110° to zero within 3 s. Moreover, the separation efficiency using different oils (such as toluene, n-decane, and hexane) in water reached (3820 LMH), with a water flux of 65.5% compared to CA membranes (2308 LMH) (Francis et al. 2023). On the other



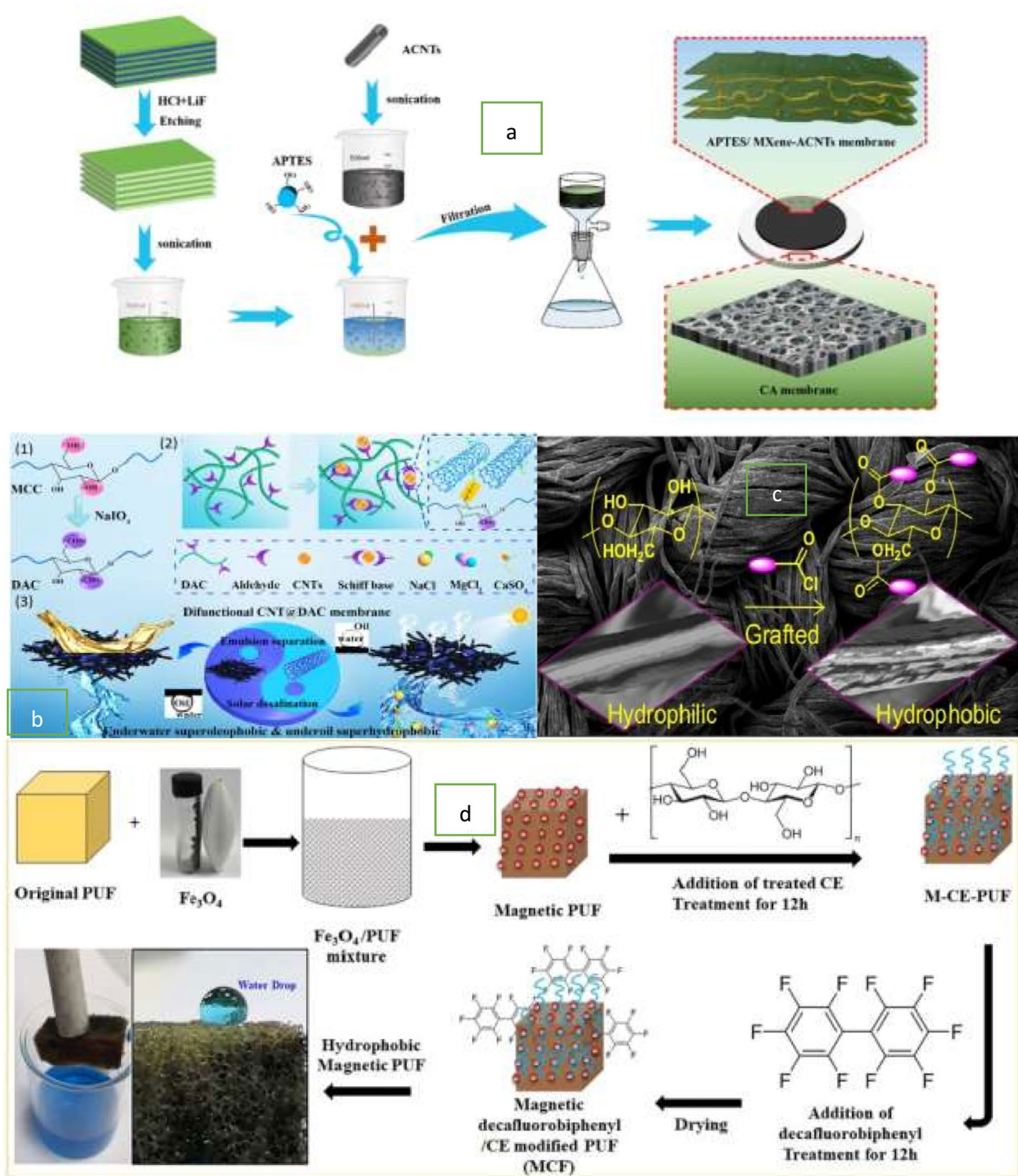
**Fig. 6** **a** BC preparation and experimental process (this figure has been adapted/reproduced from ref. Huang et al. 2023a, b with permission from Elsevier, copyright 2023), **b** fabrication of the BT@Zn membranes as a multifunctional membrane for wastewater treatment (This figure has been adapted/reproduced from ref. Wahid et al. 2022, Elsevier permission, copyright 2022), **b, c** Ni foam modified by BNC fibers (This figure has been adapted/reproduced from ref. Wang et al. 2023a, b, Springer Nature permission, copyright 2023)

hand, the super wettable (d-CA/BC@CCA) composite membrane based on regenerated electrospun (d-CA) nanofibers membrane anchored with (BC) and crosslinked with citric acid (CCA) was constructed for achieving the highly efficient oil/water purification (Ning et al. 2023). Moreover, the original d-CA membrane displayed oil/water separation efficiency higher than /BC@CCA membrane. The d-CA/BC@CCA membrane exhibited flux and separation performance of n-hexane/water emulsion without (SFE) and with (SSE) emulsifier for  $9364 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ , 98.34% and  $5479 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ , 99.39%, respectively. The reason is the remarkable hydrophilicity and reduced pore sizes formed by the unique spider-web structure (Ning et al. 2023). After 15 cycle, d-CA/BC@CCA-III membranes were illustrated a high oil/water separation efficiencies of > 99% with a slight decreased trend in separation flux. The effective separation of oil from wastewater system was investigated by prepared electrospun mates composed of (CA) and nano-zeolites (Sultana and Rahman 2022). The microfiltration process promotes the water phase to get through the CA and CA/zeolite mates and rejects the dispersed oil droplets with separation efficiency reaching 97% (Sultana and Rahman 2022).

### Cellulose-based materials membranes

In recent years, membrane-based separation technologies have had considerable prospects in oil/wastewater separation systems, where it is honored as "the water treatment technology in the twenty-first century" (Zhang et al. 2021a, b; Nabiev et al. 2023). Therefore, many researchers concern with designing and fabricating promising membranes with required properties, such as hydrophobic membranes that could only permeate oil, but water would be held (He et al. 2021). Synthesis of a series of MXene nanosheets modified with aminopropyltriethoxysilane (APTES) and amine-modified carbon nanotubes ((ACNTs) APTES/MXene-ACNTs) composite loaded on (CA) membrane by vacuum filtration was studied, as pointed in Fig. 7a (Zhang et al. 2021a, b). The composite membranes displayed a retention percentage for lubricating oil emulsion, and vegetable oil emulsion reached 99%. Moreover, APTES/MXene-ACNTs/CA (M4) provided pure water flux reaches up to  $2892.8 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$  compared to the free MXene membrane M1 ( $500.7 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ ). Moreover and for 9 cycling tests, M4 provided good antifouling property and

anti-swelling capacity (Zhang et al. 2021a, b). Nabiev et al. (2023) applied unipolar Corona discharge at a 5–25 kV voltage to treated cellulose acetate membranes for 1–5 min to separate the oil–water emulsion. The treatment enhanced the separation efficiency from 80 to 98% and the flux from 15 to  $35 \text{ dm}^3/(\text{m}^2\text{h})$ , respectively. The variation in the supra-molecular and chemical structure of the membrane could reveal the separation improvement. Another efficient type of membrane was investigated and prepared as a bifunctional composite (CNT@DAC) membrane by crosslinking of dialdehyde microcrystalline cellulose (DAC) with amino decorated multi-walled carbon nanotubes (CNTs) through Schiff base interaction for solar-driven desalination and emulsion separation, as shown in Fig. 7b (Zhu et al. 2022a, b). The CNT@DAC membrane can absorb 97% of the full solar spectrum, thus providing solar-driven evaporation efficiency, reaching 90.86% under 1.0 sun. Moreover, the membrane showed high underwater superoleophobic and underoil superhydrophobic properties for effective oil-in-water and water-in-oil emulsions segregation (Zhu et al. 2022a, b). The reusability of the prepared membrane was investigated and not showed any significant change during five separation cycles. To overcome the challenges facing surface hydrophobic modifications for oil/ water separation based on membranes, the grafting strategy forces membranes with more efficient durability (He et al. 2021). Therefore, He et al. modified the surface of the cotton fiber with stearyl chloride to improve its hydrophobicity, as cleared in Fig. 7c (He et al. 2021). Moreover, the observations demonstrated that the modified materials provide antifouling properties for various drinks and anti-washing characters over 10 cycles. Also, the WCA was kept at  $131^\circ$  after abrasion for 390 runs with a hectogram counterweight. Developing feasible and efficient separation membranes has gained great attention. Therefore, underwater superoleophobic paper-based materials with improved wet power were fabricated using economic and sustainable papermaking approaches (Xi et al. 2021). The paper sheets were modified with 1,2,3,4-butane-tetracarboxylic acid, enhancing the hydrophilicity and the crosslink of fibers (Xi et al. 2021). The treated paper sheets exhibited an oil contact angle underwater of  $167.8^\circ$  and a wet force of  $36.5 \text{ N m/g}$ . The water flux could be improved by  $25.8\text{--}4920 \text{ L m}^{-2} \text{ h}^{-1}$  by controlling the pore size average from 6.64 to  $18.9 \mu\text{m}$ . The obtaining illustrated that the smaller pore size and higher content of carboxyl-based material improve the oil retention (99.3%) even for submicron emulsion solution. Moreover, the pores' own zigzag structure, suitable for developing surface liquid bridge and collision demulsification, causes mechanical interception effects, which will improve the separation of the oil emulsion droplets (Xi et al. 2021). Recently, a promising superhydrophobic modification of cellulose-based materials' surface on honeycomb-like ZnO was synthesized (Shi et al. 2021). This



**Fig. 7** **a** Preparation of APTES/MXene- ACNTs/CA composite membranes (This figure has been adapted/reproduced from ref. Zhang et al. 2021a, b, Elsevier permission, copyright 2023), **b** (1) DAC and (2) CNT@DAC. (3) Schematic diagram of the application of bifunctional membranes in the efficient oily seawater purification (This figure has been adapted/reproduced from ref. Zhu et al. 2022a, b with

permission from Elsevier, copyright 2022), **c** Facile stearoyl chloride grafted cotton filter fabric (This figure has been adapted/reproduced from ref. He et al. 2021 with permission from Springer Nature, copyright 2021), **d** for the preparation of MCF (This figure has been adapted/reproduced from ref. Alazab et al., 2022 with permission from Elsevier, copyright 2022)

work applied plasma to treat cellulosic materials (cotton and filter paper fibers), allowing a honeycomb-like structure of zinc oxide to grow on its surface. The resulting composite was modified with stearic acid to improve the super-hydrophobicity of the cotton fibers  $151^\circ$  and filter paper to  $154^\circ$ . As a result, the composite showed high stability in highly acidic or alkaline media. After 5 cycle times, the modified cellulosic materials still maintain a high oil–water separation efficiencies of  $> 90\%$  (Shi et al. 2021). Due to their efficient easiness of separation, magnetic porous materials are of focus in oil/water separation (Alazab et al. 2022).

Therefore, the fabrication of magnetic polyurethane foam (PUF) anchored with pretreated cellulose-decafluorobiphenyl (MCF) was studied, as indicated in Fig. 7d (Alazab et al. 2022). The foam exhibited excellent oil–water separation for both oils and chemical solvents. More interestingly, the absorbed oil is released by squeezing. The observations showed that the as-synthesized foam exhibited excellent absorption affinity between (9 and 32 folds) of its weight, with separation percent ( $> 97.68\%$ ) and flux  $48,750 \text{ Lm}^{-2} \text{ h}^{-1}$  for n-hexane and superior recyclability ( $> 50$  times) in oil–water separation. After 50 absorbing-squeezing cycles, MCF illustrate efficient hydrophobicity and continued floating and maintain a significant oil capacity (Alazab et al. 2022). Due to their outstanding adsorption capacities, significant specific surface area, enhanced porosity, unique framework, and hydrophobic surface, these materials have focused by many scientists (Chu et al. 2022). Electroless deposition (ELD) is applied to coat magnetic cellulose nanofiber composite with polydimethylsiloxane to prepare hydrophobic surface ( $> 130^\circ$  of water contact angle) oil–water separation (PDMS-Ni-PW) composite. This composite is based on cellulose nanofibers (delignified porous wood, PW) as a substrate, magnetic nickel (Ni) layer, and hydrophobic polydimethylsiloxane (PDMS) (Chu et al. 2022). Owing to the porosity, hydrophobicity, lipophilicity, magnetic recovery, and high cycle compressibility, the PDMS-Ni-PW composite presented remarkable oil adsorption affinity and outstanding cyclic stability, and the composite showed superior oil retention ability. (PDMS-Ni-PW) demonstrated high oil adsorption capacity, and more than 80% of the sorption capacities were retained after 200 cycles (Chu et al. 2022). From sustainability and environmental protection perspectives, scientists directed their efforts to utilize eco-friendly and low-cost materials with excellent ability to fabricate filter material (Wang et al. 2021b, a). Hence, the facile layer-by-layer (LBL) self-immobilize approach is used to load a multilayer of chitosan (CS)-(CNCs) on stainless steel mesh (SSM) (Wang et al. 2021b, a). Referring to the numerous hydrophilic groups on the CNC's surface, the multilayer of CS-CNC's deposited mesh makes its surface highly hydrated in air and oil. As a result, the treated mesh exhibited a high retention percentage (99.92%) for crude

oil/water mixtures and oil-in-water emulsions ( $> 99.58\%$ ). Moreover, the modified mesh showed suitable regeneration and chemical stability. On the other hand, the cellulose acetate membrane filtration and antifouling affinity improved by anchoring the CNCs surface with a thin layer of polydopamine (PDA) (PDA@CNCs) (Yao et al. 2021). After that, (PDA@CNCs) were mixed with a making agent and (CA) to compose a reinforced (PDA@CNCs/CA) membrane using a solution phase transformation strategy. The results showed that the introduction of PDA@CNCs can greatly enhance the filtration, antifouling, and tensile strength of the CA membrane. Additionally, at 4 wt %, loading of PDA@CNCs enhances the permeability by 33%, flux recovery by 15%, and tensile strength by 76%. Moreover, the modification by DA enhances the lipophilicity and dispersion of CNCs, the linking strength among CNC fillers and CA matrix, and the ability of the CA composite membrane (Yao et al. 2021).

## Cellulose-based materials for removal of dyes

Synthetic organic dyes are widely acting as a coloring material in various industrial activities, such as paper, leather, paint, textiles, etc. Industries of leather and textile dyeing are the more serious sectors that discharge many harmful cationic and anionic dyes during the dyeing processes, leading to water contamination. The discharge of these dyes into the environment causes negative effects due to their stubbornness, low degradability, high molecular weight, and chemical stability. Even at low concentrations, the organic dyes may cause high health risks, e.g., cancer, cell mutation, and toxicity to humans and aquatic life. The environmental pollution caused by these dyes is caused by the tanning processes and part of the wastewater released from household laundry drainage. Therefore, during the last decades, the techniques of wastewater treatment and removal of these pollutants have become an important issue (Zhou et al. 2024; Zhang et al. 2021a, b; Al-Mhyawi et al. 2023; Heidari et al. 2023; Yuan et al. 2022; Ampawan et al. 2023; Ibrahim et al. 2022). Most traditional technologies and materials in this field are limited due to their hard regeneration and high operating costs (Zhou et al. 2024). So far, research teams worldwide still direct their work to modify the common materials and develop new ones to eliminate contaminants from wastewater.

Nevertheless, many of these materials still have some drawbacks that limit scaling (Ibrahim et al. 2022). Therefore, non-toxic, low-cost, renewable materials have broad prospects and are becoming increasingly popular in wastewater treatment applications (Zhou et al. 2024). Recently, eco-friendly materials have greatly used in the field of dye removal. Dyes can be classified based on their charges in

aqueous solution into cationic, anionic, and non-ionic dyes (Zhang et al. 2021a, b). Herein, this review aims to compare the efforts of the researchers to utilize and evaluate cellulose-based materials to remove cationic and anionic dyes from aqueous media. Table 3 represents the applications of cellulose-based materials for removing organic pollutants (Zhou et al. 2024; Zhang et al. 2021a, b; Al-Mhyawi et al. 2023; Heidari et al. 2023; Ampawan et al. 2023; Ibrahim et al. 2022; Zhang et al. 2023a, b, c; Koçse et al. 2022; Abdelhamid and Mathew 2021a, b; Nan et al. 2023; Yang et al. 2022; Shi et al. 2023; Ong et al. 2023; Liu et al. 2023; Vatanpour et al. 2023; Komal et al. 2022; Ajala et al. 2022; Zhu et al. 2022a, b; Wang et al. 2023a, b; Hamidon et al. 2022; Gowriboy et al. 2022; Batool et al. 2021).

### Cationic dyes

Methylene blue (MB) dye, which is highly used in different industries due to color stability and outstanding water solubility, is one of the most common cationic dyes (Zhou et al. 2024). It has various applicability in therapeutic, food production, biomedical, and biological processes (Al-Mhyawi et al. 2023; Heidari et al. 2023). The elimination of dye contaminants from wastewater by materials derived from agricultural wastes, such as abundant plant sources, is gaining great interest (Ibrahim et al. 2022; Zhang et al. 2023a, b, c).

Eco-friendly cellulose-based magnetic adsorbents were investigated as sustainable cellulose-based adsorbents for highly efficient to (MB)-dye removal. Zhou et al. (2024) modified biomass *Juncus effusus* (JE) with citric acid through esterification followed by magnetization to produce magnetic citric acid-modified JE pith powders (M-CA/JEPP) for removal of MB-dye as illustrated in Fig. 8a. Under basic condition, the M-CA/JEPP showed high adsorption efficiency reaching 98.34% within 10 min with the high saturation capacity 293.132 mg/g. The adsorption efficiency of M-CA/JEPP was maintained at a high level more than 98% for 5 cycles. The proposed adsorption mechanism investigated in this process indicating that the sorption involved H-bonding and electrostatic adsorption in addition to the vital role of pores and the  $\pi$ - $\pi$  interactions of hexagonal skeleton of M-CA/JEPP with the benzene rings in MB-dye molecules (Zhou et al. 2024). Polydopamine-functionalized cellulose-MXene composite was prepared as an efficient biomaterial for uptaking MB (Zhang et al. 2021a, b). Self-polymerization of dopamine hydrochloride and freeze-drying were applied to prepare cellulose/MXene composite aerogel, as shown in Fig. 8b. The fabricated composite aerogel has excellent characteristics such as low density, high stability, and high porosity as well as its higher removal capacity. The combined effect of H-bonding,  $\pi$ - $\pi$  interactions, and electrostatic attractions could explain the proposed adsorption mechanism. On the other hand, the

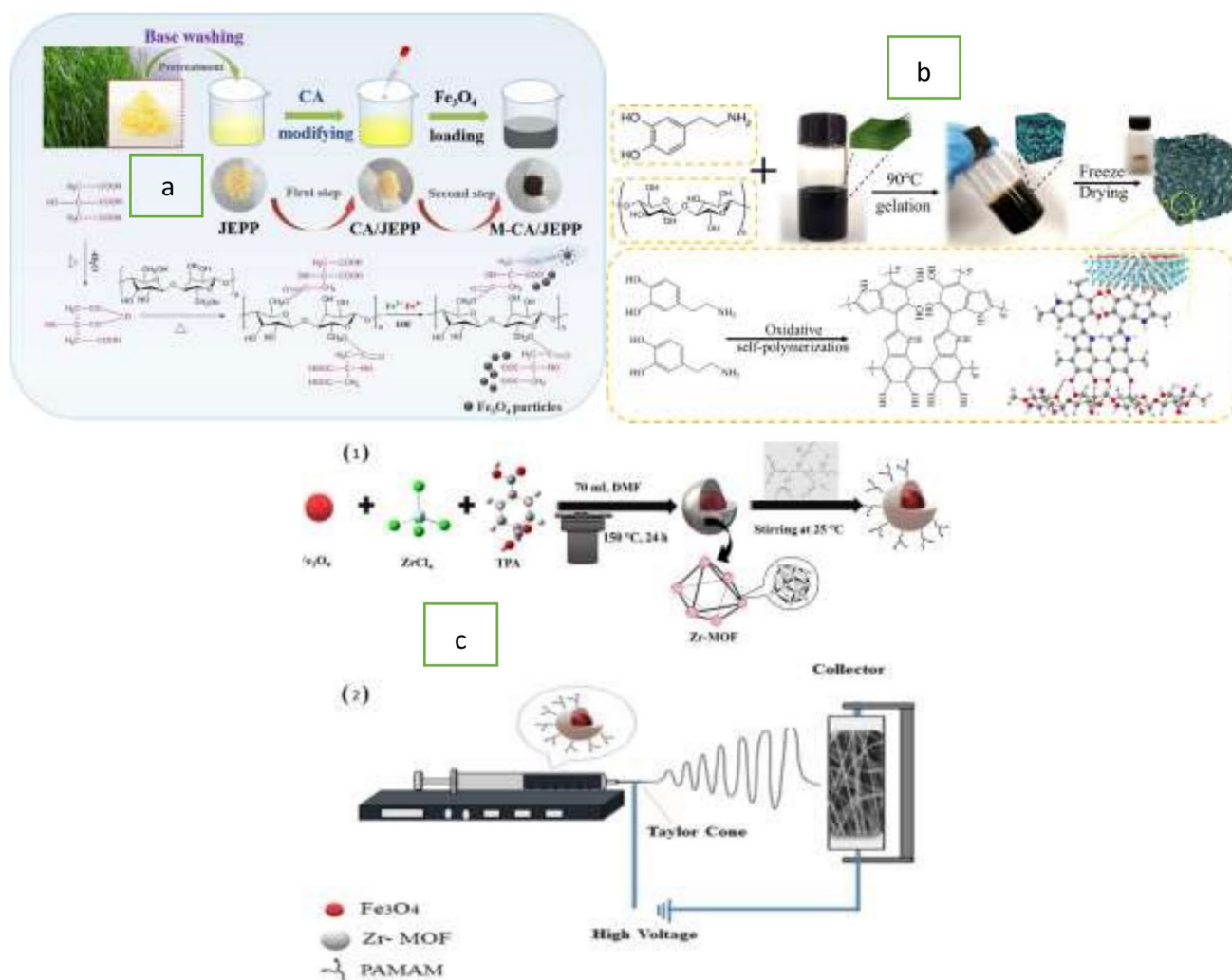
adsorption efficiencies were still remained above 84% after five cycles (Zhang et al. 2021a, b). Another bioadsorbent hydrogel (OP-PAA) to remove (MB) dye was investigated by radical polymerization of the acrylic acid (AA) and orange peel (OP) as a biomaterial rich in OH and COOH groups. The reusability of OP-PAA hydrogel was investigated and shows that the adsorption capacity retained unchanged for 10 cycles and can be regenerated at pH2 using HCl for several adsorption/desorption cycles. On the other hand, the investigated interaction mechanism of MB-dye molecules onto OP-PAA hydrogel adsorbent could be correlated more with ion exchange or electrostatic attraction (Al-Mhyawi et al. 2023). Electrospinning was also employed to prepare magnetic metal-organic framework nanofibers. There is subsequent functionalization with hyperbranched poly(amidoamine) dendrimer (PAMAM) to enhance MB-dye removal performance from aqueous media, as is shown in Fig. 8c. The experimental results illustrated that, at alkaline conditions, negative surface charges of PAMAM cause increasing removal of MB-dye molecules by electrostatic interactions. Also, after 4 cycles, the reusability investigation exhibited favorable MB-dye adsorption efficiency over 80% (Heidari et al. 2023). Yuan et al. (2022) functionalized microcrystalline cellulose (MCC) and its composites. In this research, incorporating bio-based phytic acid (PA) and condensed tannins (CT) into MCC yields PA/CT/MCC exhibiting outstanding elimination of cationic dyes. The removal mechanism of cationic dyes onto PA/CT/MCC was investigated in two steps: At the 1st step, with the flow of dyes solutions, the dyes species moves onto the surfaces of adsorbent, while at the 2nd step, H-bonding and electrostatic attractions occur between the adsorbent material and dye species (Yuan et al. 2022). A non-solvent-induced phase separation process was investigated to produce eco-friendly composite membranes based on polylactide with carboxylated cellulose microfibrers (Ampawan et al. 2023). In this work, modified cellulose microfibrers from empty fruit bunch (EFB) were mixed with maleic anhydride (MEFB) and subsequently mixed with A polylactide (PLA)/poly(butylene adipate-co-terephthalate) (PBAT) polymer blend. Adding MEFB decreases the WCA and improves the membrane porosity, as represented in Fig. 9(a, b) (Ampawan et al. 2023). In a dynamic removal approach, the permeation of pure water through the PLA/PBAT-MEFB membrane ( $1214 \text{ L m}^{-2} \text{ h}^{-1}$ ) was higher than the unmodified PLA/PBAT membrane ( $371 \text{ L m}^{-2} \text{ h}^{-1}$ ). On the other hand, 97.2% of MB was removed by PLA/PBAT-MEFB membrane, while only 58.7% was removed using neat PLA/PBAT membrane. This is attributed to the available carboxyl groups in MEFB that improve the membrane's hydrophilicity and removal ability against MB. Filtration experiments indicated that the PLA/PBAT-MEFB membrane selectively interacts MB while allowing MO to get through. Over 5 cycles of filtration, the separation of

**Table 3** Applications of cellulose-based materials for removal of dyes

Adsorbent	Modifier	Adsorbate	Adsorption capacity (mg/g)	Adsorption percent	Adsorption parameters	Reuse	Adsorption mechanism	References
MC-CA/IEPP	Magnetic citric acid	MB	293.132	98.34%		5 cycles 0.1 M HCl > 98%	Electrostatic H-bond pores $\pi$ - $\pi$ interaction	Zhou et al. (2024)
cellulose/MXene	PDA	MB	168.93		$t = 10$ h, pH 11, $T = 25$ °C, dose = 5 g/L		Electrostatic H-bond $\pi$ - $\pi$ interaction	Zhang et al. (2021a, b)
OP-PAA	PAA	MB	1930		pH 2, $t = 10$ min, dose 50 mg/100 mL	10 cycles pH2		Al-Mhyawi et al. (2023)
PAMAM	hyperbranched poly(amido amine) dendrimer	MB	940.76	94	$C_o = 21$ mg/L, dosage = 0.005 g/L, $t = 28$ min, pH 8.4, speed of stirring = 70 rpm, $T = 24$ °C	4 cycles ethanol 80.67%	Electrostatic H-bond	Heidari et al. (2023)
PLA/PBAT-ME/FB	maleic anhydride	MB MO		97.2% 58.7%	$t = 150$ min, $C_o = 10$ mg/L, pH 6	5 cycles 1 M acetic acid 96%	Electrostatic H-bond	Ampawan et al. (2023)
BC/A-zeolite	A-zeolite	MB CR 2MB/1CR 2MB/1MG		99.2% 81% (96%; 15 min, 91%; 12 h) (98.6% after 30 min)	$C_o = 10$ mg/L, dose = 120 mg, $T = 25$ °C, pH = 11	5 cycles 98%	Electrostatic H-bond n- $\pi$ interaction ionic interaction	Ibrahim et al. (2022)
HPEI-DAC	HPEI	MO	416.72		$T = 35$ °C, pH 5, $t = 5$ h,	6 cycles 0.1 mol/L NaOH 70%		Zhang et al. (2023a, b, c)
cryogels base-catalyzed CNC	Iron (II)	MO	455		pH 6, $C_o = 400$ mg/L, $t = 30$ min,			Ko'se et al. (2022)
COOH-CNF/PEI	PEI	Methyl blue Cu(II)		82.6% 69.8%	$T = 24$ h pH 5.7			Nan et al. (2023)
CNCs/PDA/ENMs/PEI	PDA/PEI	CR		99.91				Yang et al. (2022)
PEI-mica/CNFs/HPAN	PEI-mica	CBB R250 Ced EB		99.66% 98.89% 98.64%			Electrostatic H-bond	Shi et al. (2023)
CA-GO-PEI	GO-PEI	EBT MB		~90% ~80%				Ong et al. (2023)

Table 3 (continued)

Adsorbent	Modifier	Adsorbate	Adsorption capacity (mg/g)	Adsorption percent	Adsorption parameters	Reuse	Adsorption mechanism	References
NGO/ONFC/CS foam	NGO/CS	AR OG MB	416.75 300.5 14.60		pH = 4, $T = 30^{\circ}\text{C}$ , dose = 1.0 g/L, $C_o = 150\text{ mg/L}$ , $t = 12\text{ h}$	5 cycle 0.1 M NaOH ethanol– water solution (50 vol% of ethanol) Decreasing only by 7% (AR) and 12% (OG)	Electrostatic	Liu et al. (2023)
CA- ZIF-8	ZIF-8	RB5 RR 120		97 95				Vatanpour et al. (2023) Komal et al. (2022)
GO-CNF	GO	Ciprofloxacin ofloxacin	45.04 85.30		Dose = 0.5 g/L, pH 3, $t = 90\text{ min}$	5 cycles 0.2 M NaOH > 90%		
A-BSB	acid-base	Zn(II) antimicrobial		99% 100%	$t = 120\text{ min}$ , $T = 40^{\circ}\text{C}$ , dose = 0.5 g		Chemisorption	Ajala et al. (2022)
MCNC-PEI	PEI	DS	299.93		pH 4.5, $T = 25^{\circ}\text{C}$ , dose = 1 mg/mL, $C_o = 200\text{ mg/L}$ , $t = 90\text{ min}$	5 cycles 0.1 M NaOH declined by 9.9%	Electrostatic H-bond	Zhu et al. (2022a, b)
(PEI)/Zn-La (LDH)/ (CA) beads	(PEI)/Zn- La (LDH)	TC	316.76		pH 9, $t = 600\text{ min}$	5 cycles anhydrous ethanol	Electrostatic H-bonding complexation— n- $\pi$ EDA cation- $\pi$ interaction	Wang et al. (2023a, b)
CTAC-CNC/ALG@ HB	CTAC	4-chlorophenol	64.935		$C_o = 100\text{ mg/L}$ , dose = 10 g/L, $T = 30^{\circ}\text{C}$ , pH 4, $t = 6\text{ h}$	5 cycles 99% ethanol 78.65%	Electrostatic H- bonding $\pi$ - $\pi$ stacking	Hamidon et al. (2022)
Cu-MOF@CA-PES	Cu-MOF	BSA HA		85% 79%				Gowriboy et al. (2022)
PEI@CE	PEI	Polymer nanopar- ticles		> 98%				Batool et al. (2021)



**Fig. 8** Illustration of the synthesis process of **a** M-CA/JEPP (This figure has been adapted/reproduced from ref. Zhou et al. (2024, Springer Nature permission, copyright 2023), **b** composite aerogel and some chemistry mechanisms involved in the fabrication process (This figure has been adapted/reproduced from ref. Zhang et al. 2021a, b,

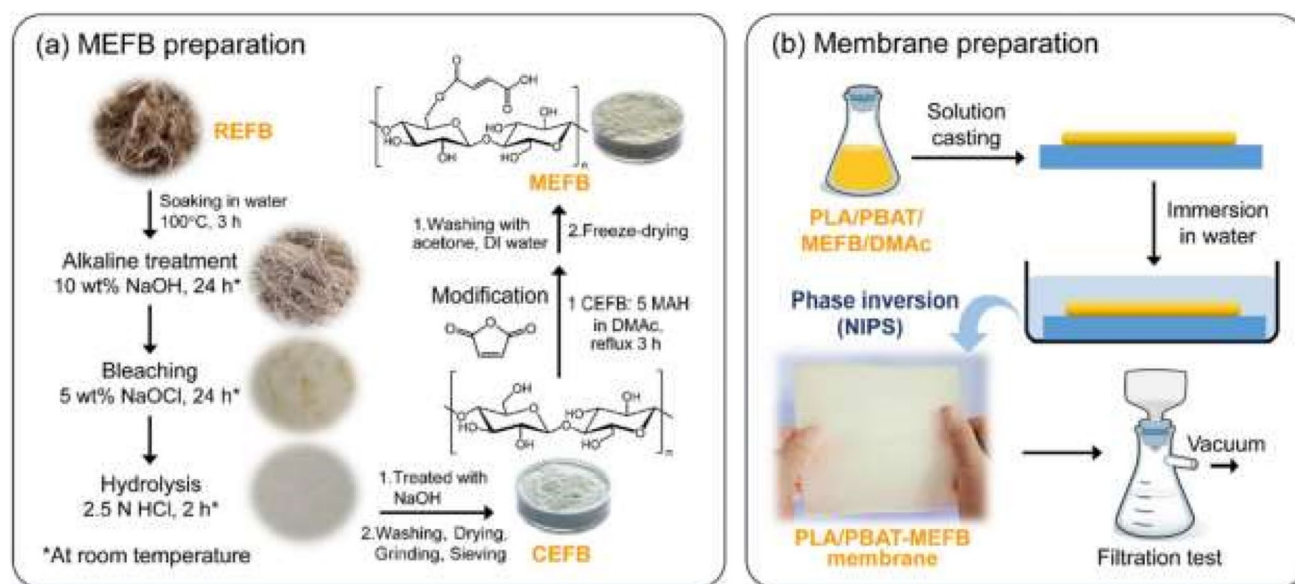
Springer Nature permission, copyright 2021, **c** (1) Fe<sub>3</sub>O<sub>4</sub>@MOF-PAMAM; (2) Schematic of the electrospinning apparatus (This figure has been adapted/reproduced from ref. Heidari et al. 2023, Springer Nature permission, copyright 2023)

dye species onto the biocomposite membrane was investigated with satisfactory separation efficiencies. Moreover, the membrane could be recovered and used for several cycles with suitable removal efficiency (Ampawan et al. 2023). Also, microcrystalline cellulose Cellets 200 were displayed as potential material to uptake MB-dye from dyed water (Suteu et al. 2019). The prepared adsorbent could be regenerated by H<sub>2</sub>SO<sub>4</sub> or CH<sub>3</sub>COOH, and this is considered as a major advantage in viable applications in environmental eco-technology.

### Anionic dyes

The efficient utilization of cellulose-based materials to adsorb anionic dyes has attracted great attention from

researchers during the last decade. Zhang et al. (2023a, b, c) oxidized cellulose derived from apple pomace to yield dialdehyde cellulose (DAC) and secondly crosslinked with hyperbranched polyethyleneimine (HPEI) to form 3D porous structure for effective MO-dye removal. The HPEI-DAC hydrogel, with excellent structure, and the (HPEI) with a high density of amine groups, possessed high adsorption capacity toward MO. Adsorption/desorption investigation of HPEI-DAC hydrogel illustrated that the adsorption capacity could reach over 70% for six cycles. The sorption of dye species onto HPEI-DAC hydrogel was explained by Langmuir sorption mechanism which supposed homogeneous sorbent surfaces with identical adsorption sites (Zhang et al. 2023a, b, c). On the other hand, the base-catalyzed CNC addition method was used to prepare CNC-modified poly(2-hydroxy



**Fig. 9** Synthesis of **a** carboxylated cellulose microfibril (MEFB) from raw empty fruit bunch (REFB) and **b** a biocomposite membrane from polylactide (PLA), poly(butylene adipate-co-terephthalate) (PBAT),

and MEFB by phase inversion (This figure has been adapted/reproduced from ref. Ampawan et al. 2023, Elsevier permission, copyright 2023)

ethyl methacrylate-glycidyl methacrylate)-[poly(HEMA-GMA)] cryogels (Ko'se et al. 2022). After that, the cryogel is electrochemically loaded with iron (II) to enhance its affinity toward anionic (MO)-dye (Ko'se et al. 2022). Also, the oxidized cellulose form was investigated as a substrate for the growth of ZIF-8 crystal (denoted as CelloZIF-8) and ZIF-L (CelloZIF-L) to remove dyes (Abdelhamid and Mathew 2021a, b). The materials showed anionic dyes' high removal ability by adsorption and degradation processes. The materials could be reused 5 times without markable loss of their ability, confirming its regeneration potential during wastewater purification. Besides the ion exchange mechanisms, other mechanisms as electrostatic interaction and  $\pi$ - $\pi$  stacking could be proposed to the investigated processes. Also, investigated Cello-ZIFs synergistically was hybrid mechanisms like catalytic hydrogenation, sorption within the pore structure, charge-specific sorption without toxic chemicals. (Abdelhamid and Mathew 2021a, b). CNFs were extracted from soybean hulls, followed by oxidation with TEMPO to produce COOH-CNFs (Nan et al. 2023).

After that, drops of zinc chloride were added to the previous suspension and left for 24 h to form hydrogel. Finally, the hydrogel is composited with PEI to produce COOH-CNF/PEI hydrogels. Modification of CNFs with polydopamine and staking on electrospun nanofiber membrane (ENMs) subsequent crosslink with PEI were studied for preparation of the nanofiltration (NF) membranes for dye removal and salt separation (Yang et al. 2022). NF-membrane was fabricated by loading PEI-functionalized mica (PEI-mica) and (CNFs) on a hydrolyzed polyacrylonitrile (HPAN) substrate

through simple vacuum filtration (Shi et al. 2023). Incorporating PEI-mica nanolayers and CNFs formed a thin and loose "brick mud" network through electrostatic interaction and H-bond. This structure allows water molecules and salt ions to flux through the channels while resisting dye molecules. The prepared PEI-mica/CNFs membrane exhibited good water flux (62.18 LMH/bar) and enhanced dye/salt separation activity, with outstanding dye resistance and low salt rejections. The experimental results demonstrated that the PEI-mica/CNFs membrane has high antifouling performance and retains stability for long term. On the other hand, adsorptive interactions, electrostatic repulsions, and size exclusion could be explained the mechanisms by which the molecules were transported through loose NF-membranes (Shi et al. 2023). In addition, the PEI-mica/CNFs composite membrane possessed suitable antifouling ability and operation stability. Many authors illustrated that membrane pretreatment applications, like microfiltration (MF), are economic and efficient (Ong et al. 2023). Therefore, modified (CA) microfiltration membranes with layer-by-layer loading of bilayers based on - Ve graphene oxide (GO) and + Ve PEI were fabricated by vacuum filtration assisted for enhanced dye rejection. The activity of 1-, 2-, and 4-bilayer-modified membranes was examined for their dye rejection of the anionic Eriochrome black T (EBT) dye and the cationic (MB) dye in a cross-flow membrane module. It was found that increasing the GO-PEI bilayers number onto the membrane increases the membrane thicknesses, and the flux of deionized (DI) water through the membranes was reduced from 4877 LMH/bar for the control (no bilayer) membrane to 2890 LMH/bar

for the 4-bilayer membrane—conversely, the performance of dye rejection of the increased. The modified membrane showed excellent rejection of anionic EBT dye (~90% rejection) compared to the cationic MB-dye (~80%), possibly due to the electrostatic repulsion among the negatively charged GO surface and anionic EBT dye (Ong et al. 2023). NGO/ONFC/CS foam based on oxidized nanofibrillated cellulose (ONFC), amino-modified graphene oxide (NGO), and chitosan (CS) produced through Schiff base crosslinking was applied for selective removal of anionic dyes. The efficiency, selectivity, and adsorption mechanisms of the investigated adsorption processes can be attributed to the electrostatic interactions toward anionic dyes and electrostatic repulsions toward cationic dyes. Recyclability studies demonstrated that the prepared materials foams were retained prominent reusability after 5 cycles (Liu et al. 2023). Also, decorated (CA)-based nanofiltration membranes with zeolitic imidazole framework-8 (ZIF-8) particles with different doses were prepared for potential water treatment and removal of anionic dyes (Vatanpour et al. 2023). Outcomes cleared that an increase in the ZIF-8 content led to a reduction in the contact angle and an increase in pure water permeation. Also, a reduced fouling ability was observed for all ZIF-8 doped membranes. Moreover, the removal efficiency of reactive black 5 dye enhanced from 95.2 to 97.7% with the addition of ZIF-8 particles. The coupling among binding sites of the membrane contaminants was reduced where ZIF-8 MOF tend to compose a compact water barrier on the membrane, and therefore, its surface became more hydrophilic surface (Vatanpour et al. 2023).

## Other organic pollutants

Rapid global industrialization in pharmaceutical and medicine industries has increased pollution of water resources, where effluents from hospital wastewaters, pharmaceutical industry, and personal sanitation products are discharged into water bodies unrestrainedly (Komal et al. 2022; Ajala et al. 2022). In this concern, a recent study focused on the elimination of ciprofloxacin and ofloxacin from an aqueous environment by using functionalized (GO) with modified (CNF) extracted from surplus biomass (Komal et al. 2022). The outcomes indicated that 20 wt. % deposited of carboxylated GO within the esterified CNF showed the highest saturation ability of 45.04 mg g<sup>-1</sup> and 85.30 mg g<sup>-1</sup> for ciprofloxacin and ofloxacin, respectively. Also, regeneration and reusability of 20% CGO-ECNF nanocomposite were investigated. The experimental results illustrated considerable adsorption efficiency for both CIP and OFL after five cycles (Komal et al. 2022). Other biosorbents were used to remediate pharmaceutical wastewaters by developing the activated sugarcane bagasse (SCB) by base (BSB),

acid (ASB), and acid–base (A-BSB) that contained Zn(II) ions and pathogens (Ajala et al. 2022). The FTIR spectra showed the existence of COOH groups in lignin and hemicellulose. The XRD showed that the chemical treatment of SCB converts the sucrose compound into cellulose. The BET analysis of the A-BSB provided the largest surface area of  $5.339 \times 10^2$  m<sup>2</sup>/g, pore volume  $2.722 \times 10^{-1}$  cc/g, and lowest pore size of 2.105 nm. Thus, A-BSB showed the highest removal, 99% of Zn(II) ions, and antimicrobial capacity of 100%. Diclofenac sodium (DS), one of the most used non-steroidal anti-inflammatory drugs worldwide, is often found water streams. It is ecotoxic, even at low concentrations (Zhu et al. 2022a, b). Polyethyleneimine-functionalized magnetic cellulose nanocrystals were fabricated as low-cost materials that can effectively and easily adsorb diclofenac sodium from wastewater bodies (Zhu et al. 2022a, b). This study functionalized magnetic cellulose nanocrystal (MCNC) with a silane coupling agent as a linker. After that, it anchored with PEI to prepare MCNC-PEI, which possessed adsorption ability of 299.93 mg/g with decreased saturation affinity by 9.9% after 5 cycles reuse and has excellent recyclability (Zhu et al. 2022a, b).

The over-reliance on tetracycline antibiotics (TC) in various medical fields has hazardous effects on the safety of the ecological systems (Wang et al. 2023a, b). Therefore, effective materials and the utilization of appropriate techniques to treat TC wastewater have always been one of the long-term global challenges. Different technologies and biomaterials have been investigated and developed to remove TC-like pollutants in the aqueous solutions (Wang et al. 2023a, b). A new (PEI)/Zn-La (LDH)/(CA) beads were fabricated with cellular interconnected network to strengthen the TC removal. The 10%PEI- 0.8LDH/CA beads reached the highest removal capacity of TC 316.76 mg/g. Reusability and regeneration study demonstrated that the sorption capacities reached to 88.88 mg g<sup>-1</sup> and remained unchanged when bead adsorbents were reused for the 5 cycles (Wang et al. 2023a, b). Another commonly detected pollutant in water streams is 4-chlorophenol, a phenolic endocrinedisrupting chemical known for its carcinogenicity and high toxicity (Hamidon et al. 2022). Anchored cellulose nanocrystals with cetyltrimethylammonium chloride (CTAC) and embedded onto the backbone of alginate were investigated for preparation of sustainable porous hydrogel beads (CTACCNC/ALG@HB) for adsorption of 4-chlorophenol with maximum adsorption capacity 64.935 mg g<sup>-1</sup>. The adsorbent hydrogel beads illustrated high reusability performance with the same adsorption capacity after 5 regeneration cycles (Hamidon et al. 2022).

4-Nitrophenol (4-NP) is one of the most hazardous aromatic compounds in different applications and industries (Yu et al. 2021b, a). Recently, grafted cellulose nanofibril with 3-dimensional PEI (CNF-PEI) was used to produce porous aerogel as a substrate for platinum (Pt) NPs to treat

4-Nitrophenol (4-NP) from wastewaters (Yu et al. 2021b, a). The Pt ions first adsorbed over amine groups of PEI and afterward reduced with NaBH<sub>4</sub> for in situ growth of Pt NPs. The aerogel exhibited excellent catalytic ability to convert 4-NP with rapid reaction kinetics ( $k = 0.12 \text{ min}^{-1}$ ). The reusability of Pt-loaded CNF aerogel showed notable efficiency as a high potential practical recyclable environmental catalyst with facile recovery for 5 cycles (Yu et al. 2021b, a).

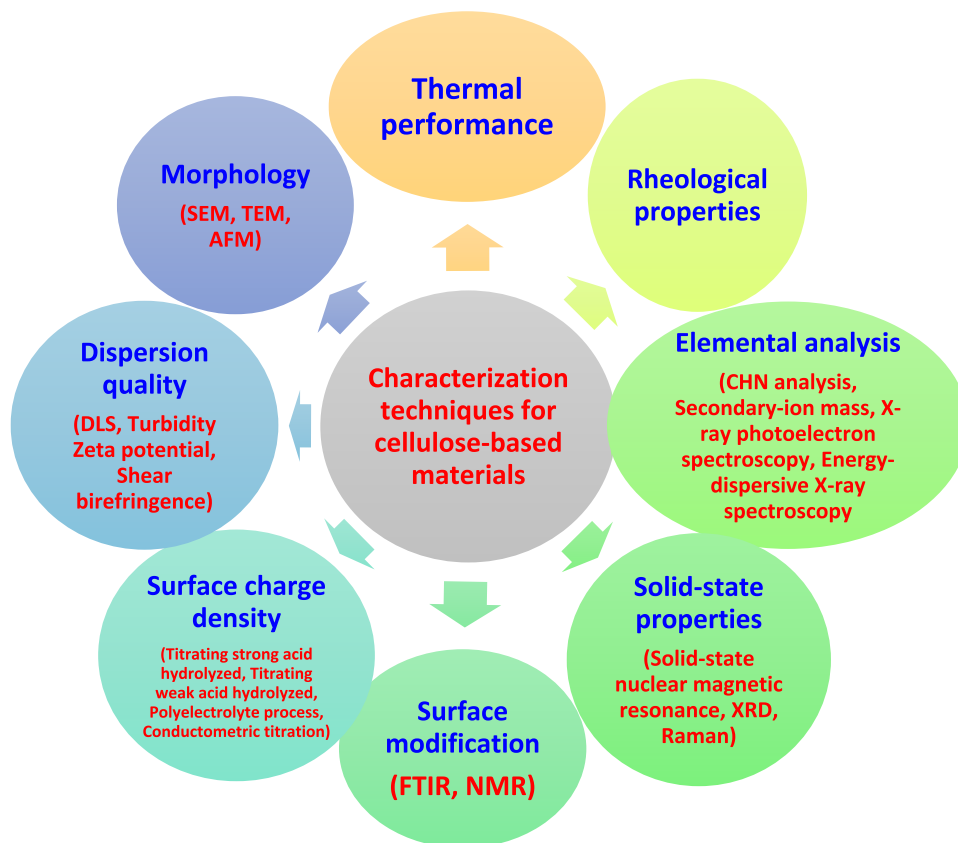
Other cellulose-based materials were prepared to improve the rejection rate for bovine sirup albumin (BSA) and humic acid (HA) by preparing hydrophilic Cu-MOF using a solvothermal strategy and blending with a (CA)/polyether sulfone (PES) polymer to produce (Cu-MOF@CA-PES) membrane via casting solution approach (Gowriboy et al. 2022). In addition, the grafting of the Cu-MOF membrane material also showed outstanding antibacterial effects against *Pseudomonas aeruginosa*, *E. coli*, *Enterococcus faecalis*, and *Staphylococcus aureus* for gram-positive and gram-negative. Batool and Valiyaveetil (2021) grafted the surface of cellulose fibers with PEI using GA as a linker for polymer nanoparticle removal from spiked water samples. The PEI@CE fibers can remove (> 98%) polymer nanoparticles within 30 min. The PEI@CE adsorbent developed was efficient, stable, and renewable to remove wide range of contaminants from wastewater (Batool and Valiyaveetil 2021).

Also, cellulose acetate/nanodiamond (CA/ND-COOH) nanocomposite membrane was fabricated as a high performance and excellent antibiofouling with high porosity properties to treat pharmaceutical wastewater in membranes bioreactors process (MBR) (Etemadi et al. 2017).

### Advantages and disadvantages of some applied characterization techniques

The characterization methods are very essential for cellulose-based materials and should be carried out before used to determine its suitability for specific applications. Some characterization techniques used for analysis of cellulose-based materials were summarized and are represented in Fig. 10. Specific surface area, aspect ratio, shape, dimensions, thermal stability, rheological behavior, crystalline structure, surface charge density, purity, and dispersion quality are the main structural characteristics for cellulose-based materials and critical to understand their role when developing cellulose materials (Zhu et al. 2021; Nagarajan et al. 2021) where the applications of cellulose-based materials have been governed in its amorphous and crystalline structures (Salem et al. 2023). Although such enabling technologies can aid in the assessment and evaluation of cellulose-based nanomaterial structural, their applications have limitations

**Fig. 10** Schematic illustration the characterization techniques for cellulose-based materials



**Table 4** Some strengths and limitations of various characterization techniques applied in the characterization of the cellulose-based materials

Technique	Strengths	Limitations
Dynamic light scattering (DLS)	<p>Gives a hydrodynamic “apparent particle size”</p> <p>Calculated coefficient is used to measure the hydrodynamic diameter (<math>H_d</math>)</p> <p>Describe the average weight and light scattering scales with the diameter of particles with power 6 (<math>d^6</math>)</p>	<p>Cannot provide accurate information regarding particles lengths or cross-sections of cellulose nanocrystals (CNCs)</p> <p>Difficulty to measure cellulose nanofibers (CNFs) suspensions</p> <p>Salt can also affect the apparent particle size</p> <p>At high CNF concentrations, multiple scattering events and potentially promote particle–particle interaction and aggregations</p> <p>Multiple peaks and inaccurate data may appear at low CNFs and CNCs concentrations</p> <p>Small particle can be entirely obscured by large particle or impurities</p> <p>Used only for assessing the colloidal stabilities</p> <p>Samples should be stable, degassed, uniform, and repeated 3 times for reliable measurement</p> <p>Turbidity changes could be linked with kinetic effects and multiple physical phenomena</p> <p>To obtain an accurate zeta potential values, some salts are added into the nanocellulose suspensions during the experiments</p> <p>Various parameters such as pH, impurities, salts, and temperature of cellulose nanosuspensions can affects zeta potential values</p> <p>No birefringence is observed with a poor dispersion</p> <p>No shear birefringence is observed above a threshold aggregate size</p>
Turbidity	<p>Assess the dispersion of cellulose nanomaterials (CNMs)</p> <p>Can be measured by UV–Vis spectrometer or Turbiscant equipment's</p>	
Zeta potential	<p>Measure the electrophoretic mobility (mm/s) of the particles</p>	
Shear birefringence	<p>Used to measure the dispersion qualities (optical/anisotropic properties) of cellulose nanomaterials suspensions</p> <p>Used as qualitative measurement to know about the dispersion qualities of cellulose nanomaterials</p>	
FTIR	<p>Provides information on molecular vibrations</p> <p>Currently used FTIR spectrometers have high signal-to-noise ratios and high spectral resolutions</p>	<p>Previous FTIR instruments were dispersive</p> <p>Other techniques can be used to obtain quantitative information</p> <p>Careful drying KBr powder and the samples of CNMs</p>
NMR	<p>One of the most powerful techniques to analyze and qualify the surface modification CNMs</p>	<p>Compared to the sensitivities of XPS or FTIR, NMR has relative low response</p>
XRD	<p>Applicable to all cellulose-based materials, such as CNFs and CNCs</p> <p>Provide qualitative and quantitative chemical structural behavior of cellulose nanomaterials</p> <p>Determine precise lattice parameters and the compositional ratio of cellulose</p> <p>Intense diffraction patterns can be obtained by using a very small amounts of samples</p> <p>Abilities to examine single fiber without needing for sectioning</p>	<p>To assess the amorphous and crystalline content in the cellulose-based materials Segal peak height method is not suitable</p> <p>Results are averaged over time and space</p> <p>Not provide a dynamic visualization of the cellulosic structures</p> <p>Patterns of organic substances (such as cellulosic materials) observed for a very short time</p> <p>Limited to evaluate the preservation of crystallinity</p>
TEM	<p>Most used technique to characterize nanoparticles</p> <p>Qualitative and quantitative method</p> <p>Estimation the particle size distributions and morphology of CNC nanoparticles</p> <p>Magnified images contrast are almost 5 times &gt; SEM</p> <p>Concentrations for TEM range around 0.01–0.50 mg/mL</p>	<p>Factors of sample preparation regarding the contrast and the dispersibility CNMs are invisible and not enhancing the resolution and contrast</p> <p>For CNC nanoparticles, allows crystallography diffraction experiment to visualize only 2D distributions</p>

**Table 4** (continued)

Technique	Strengths	Limitations
Raman	<p>Samples of both dry and wet forms are analyzed</p> <p>Used to analyze and characterize CNMs</p> <p>Utilized to obtain quantitative information</p> <p>Utilized as a qualitative and comparative tool</p> <p>Critical to determine the degree of crystallinity</p> <p>Study the conformation of purified cellulose,</p>	<p>O–H stretching vibrations is weak</p> <p>Molecules must have polarizable dipoles</p> <p>Water bands have weak intensities</p> <p>Limited when it comes to native lignocellulosic or cellulose biomass</p>

and potential pitfalls if not conducted with attention and careful judgment. (Salem et al. 2023). Therefore, this section briefly outlines and highlights the strengths and limitations of these techniques. Due to the existence of various methods and techniques in this field which there is no space to evaluate their strengths and limitations, we will display the more common techniques in this part. Some strengths and limitations of various characterization techniques were summarized and are represented in Table 4 (Foster et al. 2018; Rongpipi et al. 2019; Nagarajan et al. 2021; Shojaeiarani et al. 2021; Dassanayake et al. 2023).

## Conclusion and future perspective

The present review covers important works published recently in the literature which discussed the preparation of cellulose-based materials and their applications for wastewater remediation from different types of pollutants. Recently, cellulose-based materials have attracted scientists' interest due to their excellent advantages: economical, green, available in large amounts, low toxicity, and biodegradable. We presented the extraction of cellulose materials from their natural resources, utilizing natural cellulose materials like cotton, cellulose acetate in nanofiber, and preparation of bacterial cellulose. 3D porous materials different structures of these materials were shown, e.g., 3D porous materials, beads, membranes, etc. Although these cellulosic materials show low activity, they are considered a suitable support for the other materials. The attractive characterization of cellulose-based materials such as biocapabilities and biodegradation and biocompatibility is attracted widespread applications in wastewater treatment from different types of pollutants and oil/water separation techniques due to the ability to improve the surface properties and separation capacities of such materials via various fabrication techniques. Therefore, a modification step is essential for enhancing these materials' affinity toward different pollutants with other active ones. This activation step showed/confirmed enhancement in the produced cellulose materials toward inorganic, oil, and organic pollutants in wastewater. On the other hand, the structure of the fabricated cellulosic-based materials has also gained large attention where it plays vital roles in the field of wastewater remediation. In light of this track, the fabrication of these cellulosic materials with promising structure increases the removal efficiency, sorption mechanism, mechanical stabilities, pore size, surface modification, etc.; it requires and needs further research works to enhance such properties.

Even though the last progresses have been achieved in the modifications and applications of cellulosic-based materials for wastewater remediation, many challenges still remained and required further efforts should be done. One the main

challenges facing manufacturing CNCs nanocomposites is the achieving the homogeneity dispersion of CNCs in polymers matrices and the defects that accompany it, especially hydrophobic ones. Therefore, extensive researches are still required in this field. Another significant challenge that limits the advancement and utilization of cellulosic-based materials in wastewater remediation is the softening and swelling of these materials upon exposure to water/wastewater. Therefore, further works are needed to enhance the stabilities of cellulosic-based materials and improve its water resistances by various physical and chemical modifications. Some of the current extraction methods of cellulose nanocrystals from raw materials are still not well documented and undesirable due to low yields of cellulose nanocrystals, complicated equipment setup, and cost-effectiveness. Thus, the research gap should be supplemented with sufficient data through more experimental works using other cellulose nanocrystals isolation methods. Therefore, this research gap should be bridged by more works to investigate more suitable isolation methods (Abolore et al. (2024), Abu Rub et al. (2023), Ibrahim et al. (2024), Khan et al. (2020), Mahalakshmi et al. (2021)). Also, the treatment and purification processes of wastewater by cellulose-based materials membranes have occasionally some challenges such as fouling of membranes, coating techniques, higher energy consumptions, environmental management of processes effluents, cost of machines and equipment's utilized in harsh conditions. All of these challenges and others require constructive and effective collaborations between scientists, industrialists, and the governments worldwide to investigate better and eco-friendly technologies for wastewater remediation by the promising cellulosic-based materials where the dire need to develop of technologies and eco-friendly materials of suitable characteristics such as efficient and versatile, greener, widely availability, simple to use, and cost-effectiveness is still one of the greatest challenges presently facing societies.

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## Declarations

**Conflict of interest** The authors declare that they have no conflict of interest.

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