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A review of nanocellulose adsorptive membrane as multifunctional wastewater treatment

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ABSTRACT

Dyes, inorganic and organic solvents, heavy metals and oils represent a substantial danger to water supplies, which is a major global problem. Advanced research and development in the manufacture of green-adsorptive membranes as well as simple operation, high separation efficiency, low energy consumption, eco-friendly and affordable cost have led the way to the development of sophisticated treatments for water remediation. To date, nanocellulose has been extensively investigated as excellent biomaterials in membrane filtration due to their exceptional properties such as large specific surface area, anti-fouling behaviour, high aspect ratio, high thermal resistance, outstanding mechanical properties, biodegradability and biocompatibility. The large surface area of nanocellulose contains a large number of free hydroxyl groups, which are easily modified and functionalized has been discussed. In addition, recent progresses in the application of modified nanocellulose for heavy metal removal, oily water separation and dye extractions are surveyed, since they are potentially useful as adsorbents in the filtration membrane to enhance its performance.

1. Introduction

Water pollution is a major concern in most developing nations that dealing with a severe issue that has a significant influence on human health, environment and the aquatic ecology. According to the World Health Organization (WHO), half of the world's population will be living in water-stressed areas by 2025 (WHO/UNICEF, 2019). It is a big issue that must be handled in order to maintain the well-being of the people and ecosystem. Wastewater treatment for the recovery of water, minerals, or energy is becoming a successful way to overcome this problem. Saturated salts, heavy metals, organic compounds, pharmaceuticals, oil emulsions, dyes, and even bacteria are the most contaminants in wastewater (Abouzeid, Khiari, El-Wakil, & Dufresne, 2019; Tan, Ooi, & Leo, 2020; Zhang et al., 2020). A number of scientific researches are being conducted to eliminate these pollutants in wastewater such as adsorption, filtration, ion exchange, coagulation, precipitation, electrolysis, electrodialysis and reverse osmosis (Almeida, Oliveira, Fernandes, Godinho, & Canejo, 2020; Baruah et al., 2020; Batool & Valiyaveettil, 2021; Noor et al., 2022). Adsorption and filtration increasingly gained popularity among these methods because to its ease of use, high economic value, removal of pollutants at low concentrations and high efficiency (Khulbe & Matsuura, 2018) Unfortunately, most of these techniques need further treatment because of their prohibitively expensive and low removal efficiencies (Tshikovhi, Mishra, & Mishra, 2020). The exploration of materials that enhance high separation efficiency, low energy consumption, affordable cost, simple operation, and environment friendly is highly needed.

In recent years, nanotechnology has introduced a number of innovative nanocellulose-based materials that have shown promise in wastewater treatment (Choudhury, Sahoo, & Gohil, 2020; Dutt, Hanif, Nadeem, & Bhatti, 2020; Tshikovhi et al., 2020). Subsequently, nanocellulose emerged as a green potential adsorbent for removing various contaminants from wastewater due to worldwide environmental issues and its exceptional binding and adsorbing capacities (Trache et al., 2020). Nanocellulose (NC) is a cellulosic material that are recovered or separated from native cellulose found in plants, animals, or microbes that have at least single dimension in the nanoscale range with outstanding features such as available, low cost, high hydrophilicity, surface area, aspect ratio, mechanical strength, and stability (Lin & Dufresne, 2014; Liu, Liu, & Shen, 2021; Mateo, Peinado, Morillas-

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Review





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Gutiérrez, La Rubia, & Moya, 2021). Because of their appealing and great properties such as abundant, renewable, biodegradable, and biocompatible materials, NC are gaining increasing interest (Tienne, Santos, de Fátima, & Marques, 2020; Shaghaleh, Xu, & Wang, 2018; Trache et al., 2020; Vismara et al., 2021). There are three main categorizes of nanocellulose depend on nanocellulose's isolation technique, structure, size, and orientation (Dhali, Ghasemlou, Daver, Cass, & Adhikari, 2021). Moreover, the surface of nanocellulose contains a large number of free hydroxyl groups, which are easily modified by some functional groups to increase their performance (Gopakumar, Manna, Pasquini, Thomas, & Grohens, 2018).

The process of nanocellulose modification with various types of functional groups makes it one of the most widely used materials as an adsorptive and separative materials depending on the class of pollutants of wastewater (Mahfoudhi & Boufi, 2017). In other hand, nanocellulose can be used as pore former whereas the interaction with the polymer matrix form porous structure enhance the water filtration (Shamsuddin, Abdullah, & Othaman, 2013). The combination of excellent adsorption and filtration properties of nanocellululose aided in the development of novel biomaterial as dual-functional membranes for pollutants removal in wastewater. Due to the cost, biodegradability, eco-friendly, availability, and performance attributes, nanocellulose as adsorptive membrane gained substantial attention (Qalyoubi, Al-Othman, & Al-Asheh, 2021). The removal contaminants such as dyes, heavy metals, organic compounds and oils from wastewater by nanocellulose-based membrane adsorption has proven by many researchers (Cheng, Ye, Chang, & Zhang, 2017; Huang, Zhan, Li, Tian, & Chang, 2019; Omran et al., 2021; Sharma, Shahnaz, Subbiah, & Narayanasamy, 2020; Sorriaux, Sorieul, & Chen, 2021; Zhan, Zuo, Tao, & Chang, 2018).

This study aims to review and critically evaluating this growing area of research by exploring nanocellulose characterization, isolation methods, and modifications as well as the effectiveness in the environmental remediation for wastewater treatment. The applications of modified nanocellulose as adsorptive and separative materials for wastewater treatment has been surveyed and discussed.

2. Nanocellulose sources

The most abundant sustainable organic substance on earth is cellulose (Seddigi et al., 2021). Plants (softwood and hardwood), agricultural wastes, microorganisms, and animals are all main source of cellulose (Isik, Sardon, & Mecerreyes, 2014; Klemm, Heublein, Fink, & Bohn, 2005). According to Dufresne (2019), a tree generates roughly 10 g of cellulose per day, and the worldwide cellulose output is estimated to be 1.5×10^{12} t/year. Cellulose, $(C_6 H_{10} O_5)_n$ is a naturally formed raw material that was primarily discovered as a component of plant cell walls (Sjahro et al., 2021). It coexists in the wall with other lignocellulosic components, particularly hemicellulose and lignin as seen in Fig. 1. Softwood have a cellulose level of 30-75%, whereas hardwood has a cellulose content of 40-50% (Seddiqi et al., 2021). The most common softwood and agricultural wastes are rice husk (de Oliveira et al., 2017; Shahi, Wang, Adhikari, Min, & Rangari, 2021; Yunus, 2019), oil palm empty fruit bunch (OPEFB) (Mazlan et al., 2020; Mohtar, Tengku Malim Busu, Md Noor, Shaari, & Mat, 2017; Yimlamai, Choorit, Chisti, & Prasertsan, 2021), cotton (Ibrahim, Al-Khateeb, Hussin, & Al-Obaidi, 2015; Li, Zhou, et al., 2021; Zhai et al., 2021; Zhou, Fu, Liu, Gu, & Guo, 2021), sugar palm fibre (Ilyas, Sapuan, Ishak, & Zainudin, 2017; Ilyas, Sapuan, & Ishak, 2018; Ilyas, Sapuan, Ibrahim, et al., 2019; Nazrin, Sapuan, Zuhri, Tawakkal, & Ilyas, 2021, 2022; Sanyang, Sapuan, Jawaid, Ishak, & Sahari, 2016; Syafiq, Sapuan, Zuhri, Othman, & Ilyas, 2022), Agave gigantea (Edi Syafri et al., 2022), sugarcane bagasse (Nkosivele, Mzimela, Linganiso, Revaprasadu, & Motaung, 2018; Somvanshi & Gope, 2021; Zhang et al., 2016), bamboo (Kwak, Lee, Lee, & Jin, 2018; Yang et al., 2020; Yuan, Wei, & Wen, 2019), pomelo peel (Liu, Liu, Ibrahim, Yang, & Huang, 2018; Mat Zain, 2014; Tang et al., 2020), lemongrass (Kumari, Raza, & Meena, 2021; Mishra et al., 2018) lemon



Fig. 1. The lignocellulosic biomass cell wall structure (Bertella & Luterbacher, 2020).

seed (Zhang et al., 2020), wheat straw (Qasim et al., 2020; Qi et al., 2020; Yu et al., 2021), ginger (Abral et al., 2020a; Abral et al., 2020b), water hyacinth (Syafri et al., 2019), barley straw (Fortunati et al., 2016; Lara-Serrano, Morales-delaRosa, Campos-Martín, & Fierro, 2019), pineapple leaf (Gaba et al., 2021; Shamsuddin et al., 2013), and kenaf (Narkpiban & Poonsawat, 2020; Nurul Atiqah et al., 2019; Sabaruddin et al., 2020). Poplar (Chen, Xiao, Shi, & Cai, 2018), pine (Mormann, 2003), aspen wood (Jonasson, Bünder, Niittylä, & Oksman, 2020) and balsa (Marwanto et al., 2021) are some of the wood feedstocks used.

In contrast to plants, bacteria cellulose is produced in virtually pure form, up to 90% without presence of lignin and hemicellulose by the terminal complex of bacteria. Because of its great purity, BC is regarded as a non-cytotoxic, non-genotoxic, and highly biocompatible substance, drawing interest in a variety of fields, including medicine (Gorgieva & Trček, 2019). On the other hand, bacterial and plant cellulose exhibit similar crystallinity, water retention, tensile strength, and biocompatibility (Khalil et al., 2020). The most often employed bacterium for generating bacteria cellulose is Komagataeibacter xylinus (previously Acetobacter xylinum) (Swingler et al., 2021). Meanwhile, tunicates are a kind of invertebrate that lives in large numbers in the seas and is the only known animal source of cellulose (Zhao & Li, 2014). About 60% of cellulose and 27% nitrogen-containing components are found in dry tunics. The amount of cellulose, the degree of crystallinity, and the size of the crystalline domains vary depending on the cellulose sources shown in Fig. 2.

Shrinkage of cellulose to nanoscale ranges is expected to spur the development of novel nanomaterials, with the goal of broadening their uses in the field of functional nanomaterials (Dutt et al., 2020; Tshikovhi et al., 2020). The transforming cellulose into nanocellulose by different physicochemical approaches has potential scope of this nanomaterial in wastewater treatment (Mbakop, Nthunya, & Onyango, 2021; Trache et al., 2020). Nanocellulose can be extracted from cellulose materials by isolation process such as acid hydrolysis, mechanical disintegration, and enzymatic delignification (Mautner, 2020; Mbakop et al., 2021;

Zielińska, Szentner, Waśkiewicz, & Borysiak, 2021).

3. Nanocelullulose structure and properties

Nanocellulose is made up of hydrogen, oxygen and carbon in a linear form of homopolysaccharide made up of β -D-glucopyranose units connected by β -1,4 glycosidic bond (Fig. 3) (Krause Bierhalz, 2021) in nanoscale ranges. Each anhydrous glucose segment has one main hydroxyl group at C6 and two secondary hydroxyl groups at C2 and C3. Both crystalline and amorphous portions of nanocellulose are orderly structured and maintained by hydrogen bonds and van der Waals interactions between and among molecules in the aggregated structure (Pei, Wang, Tang, & Kaplan, 2021).

Nanocellulose is the most abundant polysaccharide on the planet that being used as a desirable material for a variety of reasons, including its low weight, ubiquitous nature, biodegradability, cost-effectiveness, renewable, less energy, large surface area and interesting specific properties, as well as the fact that it is waste biomass with good mechanical properties (Bhat et al., 2019; Ilyas, Sapuan, Atiqah, et al., 2020; Ilyas, Sapuan, Ishak, et al., 2019). Nanocellulose materials can be classified into three groups based on their isolation methods, which are



Fig. 3. Structure of nanocellulose.



Fig. 2. Comparison of the cellulose content derived from different resources along with the morphology and crystallinity of nanocellulose (Dhali et al., 2021).

cellulose nano-fibrils (CNF), cellulose nanocrystals (CNC), and bacterial nanocellulose (BC) (Mbakop et al., 2021) (Fig. 4). Both CNF and CNC are made up by top-down processing methods (Kargarzadeh, Ioelovich, Ahmad, Thomas, & Dufresne, 2017; Nasir, Hashim, Sulaiman, & Asim, 2018) while BC is made up of purest forms cellulose nanofibers released extracellularly by certain bacteria, synthesized from the bottom up (de Amorim et al., 2020).

Typically, CNF synthesized by mechanical process of fibril-like with a diameter below 100 nm and a length of few micrometres (Kargarzadeh et al., 2017). Meanwhile, CNC are usually made by acid hydrolysis produced rod-like crystals with a diameter of 10–20 nm and a length of 5–200 nm (Reshmy et al., 2021). Besides, BC made by bacterial synthesis exists in the form of ribbon-like-web with diameters of 20 nm to 100 nm, micrometer lengths, and an 80–90% unique crystallinity (Asim, 2018; Nechyporchuk, Belgacem, & Bras, 2016).

The use of nanocellulose within the polymers can affect the tear strength, abrasion resistance, adhesion properties (Ilyas, Sapuan, Ishak, et al., 2019) and could acted as a pore forming agent (Arman Alim &

Othaman, 2018). The specialties of nanocellulose for this particular application is due to their high aspect ratio and accessibility of plenty of –OH groups for the reinforcement with polymer composite (Ilyas, Sapuan, Atiqah, et al., 2020; Nazrin et al., 2020). To date, nanocellulose has been investigated as a potential bio sorbent for contaminant in wastewater (Putro, Kurniawan, Ismadji, & Ju, 2017; Salama et al., 2021). The availability of a large number of –OH groups in nanocellulose able to adsorb the dyes, heavy metals, and other contaminants in wastewater. As seen in Table 1, nanocellulose offers a number of benefits for wastewater treatment application.

4. Nanocellulose extraction methods

Isolating cellulose into nano-sized particles (nanocellulose) induced special chemical and physical properties of biomaterials (Li et al., 2017) and sparked a lot of curiosity in a variety of fields (Yassin, Gad, Ghanem, & Abdel Rehim, 2019). Various isolation procedures such as chemical, mechanical and biological treatment, which can be applied individually



Fig. 4. Types and properties of nanocellulose.

Table 1

The advantages and applications of nanocellulose in wastewater treatment.

Author (year)	Advantages of nanocellulose	Applications		
Abou-Zeid, Ali, Gawad, Kamal, Kamel, & Khiari (2021); Abouzeid, Khiari, El-Wakil, and Dufresne (2019) Baruah et al. (2020)	High mechanical properties, large surface area, biodegradable, reduce cost, environmental inertness and simple surface modification High surface area, high mechanical resistance, good	Metal cation removal Cu ²⁺ removal		
Fiol et al. (2019) Hernandez-Francisco et al. (2020) Hong et al. (2018)	stability and renewable Renewable and abundant Enhanced thermal stability, abundant, cost-effective Renewable, attractive bio- sorbent due to easy chemical	Cu(II) adsorption Pb(II) ion removal Metal ions removal		
Reshmy et al. (2022)	modification, chemical stability and hydrophilic Cost-effective, energy-efficient alternative, hydrophilicity, functionalization potential	Heavy metal remediation		
Mohammed et al. (2016)	High specific surface area, good mechanical strength, biodegradability and high	Sensing and scavenging of heavy metal ions		
Sharma, Anjana, H., and Goswami (2021)	Nano-dimension, large surface area, higher mechanical strength, thermal stability, and biocompatibility as compared	Heavy metal removal		
Bai et al. (2017, 2020)	to other biomaterials Desirable mechanical properties, hydrophilic, low cost plentiful available	Methylene blue removal		
Beh, Lim, Lew, and Lai (2020)	Specific surface area, good biodegradability, multi- functional material	Methylene blue removal		
Selkälä et al. (2019)	Readily applicable, environmentally friendly, and potentially cost-effective	Anionic dye removal		
Cruz-Tato et al. (2017)	Biodegradable, non-toxicity, cost-effective, enhance	Water purification		
Almeida et al. (2020)	Biocompatibility, recyclable, eco-friendly, hydrophilic, super-oleophobicity	Waste-oily water		
Gopakumar et al. (2018)	Affordable, sustainable, hydrophilic, inert, and stable	Wastewater treatment		
Ibrahim, Sazali, Salleh, and Zainal Abidin (2021)	Large surface area, high tensile strength, high resistivity to chemical and ease of surface	wastewater remediation and gas separation		
Liu et al. (2021)	functionalization High strength and specific surface area, pollution-free and	Wastewater treatment		
Mautner (2020)	renewable High surface area, abundance OH group, easily modified	Water treatment		
Mohammed, Grishkewich, and Tam (2018)	Excellent surface functionalization possibility, high surface area, high aspect ratio bich mochanical strength	Water treatment		
Herrera-Morales, Turley, Betancourt-Ponce, and Nicolau (2019)	Environmentally benign, stable, and have a high aspect ratio and high surface to	EOC removal		
Chen et al. (2019)	volume ratio Convenient preparation, sustainable material low cost	Bacteria removal		
Lu, Liu, Chen, and Luo (2021)	Hydrophilic, large surface area, high mechanical strength	Antibiotics removal		

or in combination used to extract nanocellulose materials with different shape, structure and applications (Mondal, 2017; Phanthong, Reubroycharoen, Hao, Xu, & Abudula, 2018). Table 2 shows the types and properties of nanocellulose on the isolation method. The isolating proposes to increase specific surface area, flexibility and mechanical qualities of fibre by facilitating the breakdown of cellulose fibers (Abdul Khalil, Bhat, & Yusra, 2012; Abouzeid et al., 2019). From previous data, various isolation methods result in different types and properties of nanocellulose (Phanthong et al., 2018).

Generally, acid hydrolysis and mechanical disintegration are widely used for nanocellulose extraction of lignocellulose biomass (Ilyas et al., 2021; Ilyas, Sapuan, Ishak, et al., 2019; Sutrisno et al., 2020; Zhang et al., 2020). In 2020, Zhang et al. extracted cellulose nanocrystals (CNC) from lemon (Citrus limon) seeds by using three different methods, which are sulfuric acid hydrolysis, ammonium persulfate oxidation, and TEMPO oxidation. According to the results, TEMPO oxidation produced a higher yield, larger size, and lower crystallinity index (CrI) than others. In comparison, ammonium persulfate oxidation yielded the highest CrI whereas sulfuric acid hydrolysis yielded the smallest size. Ilyas et al. (2018) were extracted sugar palm fibers (Arenga pinnata) nanocrystalline cellulose (SPNCC) by using sulfuric acid hydrolysis. Subsequently, sugar palm fibers nanofibrillated cellulose (SPNFC) were extracted mechanically by using high pressure homogenization (HPH) (Ilvas, Sapuan, Atigah, et al., 2020). The isolated SPNCCs and SPNFC were found to have crystallinity index (CrI) of 85.9% and 81.19% respectively.

4.1. Pretreatment

In lignocellulosic biomass, the cellulose and hemicellulose are often coated and cross-linked by lignin, which inhibits glycoside hydrolase (GHs) hydrolytic activity. As a result, pretreatment is required to remove the lignin from the feedstock to make these carbohydrates available for enzymatic hydrolysis and fermentation (Behera, Arora, Nandhagopal, & Kumar, 2014). According to Behera et al. (2014), pretreatment can reduce cellulose crystallinity, enhance porosity (accessible surface area), and improve biodegradability of biomass by removing lignin and hemicellulose. Several pretreatments have been suggested to aid fibre disintegration (Abouzeid et al., 2019). Alkali pretreatment is widely used as chemical pretreatment procedure that is based on lignin dissolution rate in an alkali solution and make the carbohydrates available used for the downstream processes (Baruah et al., 2018). Consequently, chemical pretreatment, such as (2,2,6,6-tetramethylpiperidin-1-oxyl) (TEMPO) oxidation, and enzymatic pretreatment are commonly practiced after alkali treatment to reduce energy consumption for producing CNF (Reshmy et al., 2021).

4.2. Mechanical treatment

A vigorous mechanical shearing approach that allows the gradual release of constitutive microfibrils is used in a top-down processing method made of cellulose nanofibrils (CNF) from cellulose pulp fibers (Pei et al., 2021). Refining or high shear homogenization, micro fluidization, grinding, cryo-crushing and ultrasonication are all part of the mechanical extraction process, which results in long chain of microfibrils and nanofibrils (Campos, Corrêa, Claro, & Marconcini, 2019; Salama et al., 2021). Both crystalline and amorphous domains are formed during mechanical shearing treatment that have a high aspect ratio and shear-thinning and thixotropic action in water, forming gels (Nechyporchuk et al., 2016). Typically, CNF have a diameter of below 100 nm and a length of few micrometres (Kargarzadeh et al., 2017). Kumode, Bolzon, Magalhães, and Kestur (2017) developed a balsa tree nanocellulose reinforced castor seed cake to generate "green" composites by mechanical treatment. Mechanical fibrillation was used to produce the nanofibrillar cellulose, and the integration of nanocellulose resulted in enhanced flexural modulus, swelling thickness, and water absorption as the amount of nanocellulose added increased (Kumode et al., 2017).

4.3. Acid hydrolysis

Acid hydrolysis is a method that uses strong or mild acid to dissolve the amorphous portion of cellulose fibre, producing the material with a

Table 2

The types and properties of nanocellulose on the isolation method.

Sources	Isolation Method	Types of nanocellulose	Morphology	Diameter (nm)	Crystallinity (%)	Zeta potential (mV)	References
Lemon seeds	Acid hydrolysis	CNC	Rod-like structure	12-25	69.67	-40.27	(Zhang et al., 2020)
	Ammonium persulfate oxidation	CNC	Rod-like structure	10–20	74.4	-31.27	
	TEMPO oxidation	CNC	Rod-like structure	26-42	66.14	-55.67	
Sugar palm fibre	High pressure homogenization	CNF	Thread-shape	21.37–5.5	81.19	-34.2	(Ilyas, Sapuan, Ishak, et al., 2019)
	Acid hydrolysis	CNC	Needle-like shape	9 ± 1.96	85.9	$\begin{array}{c} -61.50 \pm \\ 1.65 \end{array}$	(Ilyas, Sapuan, & Ishak, 2018)
Corn husk	Ultrasonication	CNF	Slender interconnected webs	$\begin{array}{c} 20.14 \pm \\ 4.32 \end{array}$	53.4	-24.3 ± 2.5	(Yang, Han, et al., 2017)
Corncob residue	Acid hydrolysis	CNC	Long-rod shape	6.5 ± 2.0	63.8	-14.3 ± 0.4	(Liu et al., 2016)
	TEMPO-mediated oxidation	CNF	Twisted structure	$\textbf{2.1} \pm \textbf{1.1}$	49.9	-23.1 ± 2.3	
Bacterial strain Komagateibacter xylinus	Static culture 96 h at 30 °C	BC	Denser network structure	$\begin{array}{c} \textbf{29.13} \pm \\ \textbf{6.53} \end{array}$	47.4	-44.1 ± 0.91	(Gao et al., 2020)
	Agitated culture 300 rpm at 30 °C	BC	Porous network	$\begin{array}{c} \textbf{29.51} \pm \\ \textbf{8.03} \end{array}$	22.1	-46.5 ± 1.51	
Cotton fibre	Acid hydrolysis ultrasonication	CNC	Rod-like shape	8–70	81.91	-	(Ibrahim et al., 2015)
Sisal fibre Agave gigantea	Chemical-ultrafine grinding	CNF	Individual fibril- fibril	4.07	71	-	(Edi Syafri et al., 2022)
Macaranga hypoleuca	Acid hydrolysis- ultrasonication	CNF	Interconnected web	42 ± 7.27	76.9	-	(Sutrisno, Tanpichai, & Chuangchote, 2020)

high crystallinity structure (Ibrahim et al., 2021). Cellulose nanocrystals (CNC) are usually made by acid hydrolysis distributed in water under carefully regulated temperature, duration, and acid-to-cellulosic fibre ratio conditions. According to Cheng et al. (2017), acid hydrolysis was used to create fibrous tunicate cellulose nanocrystals (TCNCs), which demonstrated a high degree of crystallinity and characteristic cholesteric liquid crystal behaviour. Concentrated acids, such as sulfuric acid, hydrochloric acid, or phosphoric acid, are often employed in this process to break hydrogen bonds, break amorphous areas of cellulose, and leave

crystalline sections intact (Dufresne, 2013).

The above studies clearly demonstrate that chemical pretreatment is a viable pretreatment technique owing to its potential to facilitate the disruption of various lignocellulosic materials (Baruah et al., 2018; Mafa, Malgas, Bhattacharya, & Rashamuse, 2020). Unfortunately, this method involves the use of harmful chemicals and the generation of effluent (Phanthong et al., 2018). Thus, the mechanical disintegration able to scale down the nanocellulose without wastewater. However, the process is energy intensive and detailed investigations are necessary to



Fig. 5. Nanocellulose surface modification techniques of adsorption based on pollutant class (Mahfoudhi & Boufi, 2017).

optimize the process parameters for high-scale applications. Therefore, novel extraction methods with simple steps which do not have wastewater are necessary to obtain properties and yield of nanocellulose that are equivalent to previous approaches.

5. Nanocellulose modification

Nanocellulose has gained a lot of interest as a biomaterial for diverse uses in environmental remediation because of its unique features such as abundant, biodegradable, low cost, high thermal and mechanical properties, large surface area, high aspect ratio, hydrophilic, and simple surface modification (Bangar et al., 2022). Additionally, the benefits of cellulose-based material are its ease of handling, reusability, eco-safety, and sustainability, as it can be made from waste biomass, following the circular economy's virtuous path. Nevertheless, hydrophilic properties result difficult to disperse evenly in any liquid or matrix. Therefore, some nanocellulose modification was necessary to overcome nanocellulose's incompatibility, poor interfacial adhesion, and difficult dispersion in a polymer matrix (Ilyas, Sapuan, Ibrahim, et al., 2020). Besides, nanocellulose's adsorption capability in adsorption process is still modest and has to be enhanced by additional modification or compounding (Qiao et al., 2021). Fig. 5 showed the nanocellulose surface modification techniques based on the pollutant class for adsorption process. The presence of reactive hydroxyl groups on the surface nanocellulose can be functionalized and changed in a number of ways (Sharma et al., 2021).

Sanguanwong et al. (2021) reported the assembly of modified cellulose nanofibers (CNF) to fibrous clay sepiolite (SEP) composite foams with TEMPO (2,2,6,6-tetramethylpiperidin-1-oxyl)-oxidized and methyltrimethoxysilane (MTMS) deposition, indicating that the maximum sorption capacity of olive and motor oils is 138 and 90 g/g, respectively. Fiol et al. (2019) studied TEMPO-oxidized functionalized cellulose nanofibers as a promising Cu (II) adsorbent for wastewater treatment. The results showed that the synthesized 3D CNF-aerogel structure is an effective sorbent for removing copper ions from aqueous solutions, and that using this structure for environmental decontamination opens up new possibilities for CNF uses.

Alsaiari et al. (2021) produced polypyrrole/nanocellulose (ppy/NC) nanocomposite by hydrolyzing rice straw cellulose with sulfuric acid and then functionalizing the nanocellulose with polypyrrole (ppy) through a polymerization process. The modified nanocomposite was interestingly utilised for Cr(VI) uptake for up to six cycles, with outstanding regeneration results. In a packed column, Fiol et al. (2021) compared Cu(II) adsorption of cellulose nanofibers with calcium alginate beads. Due to the unfavourable impacts of highly charged water and diffusion through the beads, the adsorption rate of CNFs was much greater than that of calcium alginate beads. The results reveal that by utilising a rapid, simple, and efficient chemical modification based on surface esterification, a high-performance bio-based and nanostructured cellulose material may be used as a Cu(II) adsorbent.

By combining carboxymethylation and homogenization for heavy metal ions removal (Cu²⁺), Famei et al. produce carboxymethylated CNFs (CMCNFs) with a carboxylate concentration up to 2.7 mmol/g. The influence of several experimental variables (such as pH, carboxylate content, contact duration, and starting Cu^{2+} concentration) on the Cu^{2+} removal capability of CMCNFs is studied in depth. CMCNFs have a record high equilibrium Cu^{2+} elimination capacity of 115.3 mg/g at pH 5.0, according to their adsorption performance. Same study by Hong et al. (2018) for carboxymethylated cellulose nanofibrils (CMCNFs) embedded in polyurethane foam. Well distributed modified nanocellulose (CMCNFs) embedded in PU matrices and the resulting hydrogen bonds result in a large increase in mechanical strength, which is another prerequisite of appropriate adsorbents in wastewater treatment. These findings show that in-situ integration of CMCNF into a water-born PU scaffold during polymerization is a simple, quick, and effective way for modularizing different nano-scaled adsorbents for

metal ion removal.

Through the integration of graphene oxide (GO) and trimethylolpropanetris-(2-methyl-1-aziridine) propionate (TMPTAP), Mo et al. (2021) used a facile strategy to fabricate highly elastic 3D TEMPOoxidized cellulose nanofiber (TCNF) aerogels (TCNFAs) with selective capture capacity and superfast adsorption for removing Pb(II) species. For effective removal of organic dyes from water, it consists of a microand nanoporous sponge-like system derived by thermal cross-linking among (2,2,6,6-tetramethylpiperidin-1-yl)oxyl (TEMPO)-oxidized cellulose nanofibers (TOCNF), branched polyethylenimine 25 kDa (bPEI), and citric acid (CA) (Riva, Pastori, Panozzo, Antonelli, & Punta, 2020).

Ji, Wen, Wang, Zhang, and Guo (2020) created an eco-friendly and low-cost microporous aerogel by grafting renewable cardanol-derived siloxane onto a cellulose nanofiber (CNF) framework to collect Cu(II) and organic contaminants. Hydrophobicity, which was crucial for very effective absorption of Cu(II) and oil droplets. Modified aerogels had a saturation adsorption capacity for Cu(II) of 45.6 mg/L, which is greater than the majority of published biobased adsorbents. With maximal absorption capabilities of up to 108 g/g, modified aerogel efficiently gathered different oils and organic solvents from water (chloroform). Modified aerogels are environmentally friendly and cost-effective, since they do not include any extra harmful chemicals. This allows them to considerably minimise secondary pollution and contaminated emissions to the environment after usage.

In comparison to CNCs and GO, the GO/CNCs composite can give a better possibility for [bmim][Cl] sorption because to the ease of producing 3D crumpled structures and the abundance of oxygen in the functional groups. The results revealed that the sorption kinetic was well suited by the pseudo-second-order kinetic model and the Eovlich model. The Langmuir model, with a maximum sorption capacity of 0.455 mmol/g, was found to be better characterized by the isotherm adsorption data. This study shows how to make a 3D structure adsorbent from graphene oxide and cellulose nanocrystals that has a high [bmim][Cl] adsorption capability in aqueous solution (Zhou et al., 2017). A study found that cationic-modified nanocellulose is able to adsorb nanoscale viruses. The cationic cellulose-based aerogel filter has a viral removal capacity of 99.9% for MS2 and 93.6% for Qbeta at pH = 7.0, while at pH = 3.0, desorption of largely intact viruses occurs (Watts, Maniura-Weber, Siqueira, & Salentinig, 2021).

For bacterial cellulose (BC), modification and incorporation of other molecules are still required to expand its range of applications although BC has many advantageous properties for various applications (Blanco Parte et al., 2020). BC lack effective capabilities to trigger initial cell attachment and control over porosity, and it degrades slowly, among other things (Gorgieva & Trček, 2019). Therefore, some versatile methods such as in situ and ex situ has been done to modify BC chemically (chemical structure and functional group) and physically (change in porosity, crystallinity, and fibre density). Ex situ modifications are accomplished through the modification of culture media, carbon sources, and the addition of other materials, whereas in situ modifications are accomplished through the chemical and physical treatment of formed BC (Blanco Parte et al., 2020; Gorgieva & Trček, 2019). Besides, according Blanco Parte et al. (2020), the oxidation of cellulose's hydroxyl groups is a common modification, with the most common method being 2,2,6,6-tetramethyl-1-piperidinyloxyl (TEMPO). TEMPO directly targets primary hydroxyls by converting them into carboxylated cellulose (CBC), which can then be used to covalently anchor functional molecules.

Dórame-Miranda et al. (2019) conduct experiment to produce bacterial cellulose by *Gluconacetobacter entanii* in a static culture medium with pecan nutshell as the carbon source and saccharose as the control. According to the findings, the pecan nutshell could be used as a carbon source for BC production, with a cellulose yield of 2.816 ± 0.040 g/L after 28 days. Hence, the BC could be used to develop bio-composites. Chaabane et al. (2020) used a multi-step procedure to develop novel magnetic materials containing *tetraaza macrocyclic Schiff* base bacterial cellulose ligands and magnetite nanoparticles (Fe₃O₄NPs) for antimicrobial and cytotoxic activities as well as chemotherapy in cancer treatment. The ethylenediamine (EDA) and benzil (Bzl) were used to chemically modify 2,3-dialdehyde bacterial cellulose (DABC) in the presence of ferrous ions. Magnetite nanoparticles (Fe₃O₄NPs) were then synthesized using a co-precipitation method within the complex [Fe (DABC-EDA-Bzl)Cl₂. Anti-tumor study showed that the magnetic [Fe₃O₄NP-INS-(DABC-EDA-Bzl)] material efficiently prevent the development of the CT26 tumor model in BALB/c mice compared to other resulting materials over the period of the experiment and can be used for effective drug delivery in nanomedicine.

In 2018, Badshah et al. (2018) conducted experiment to develop surface modified BC matrices loaded with model drugs chosen for their aqueous solubility. The results showed most of the drug was released in 0.5–3 h and 0.25–0.5 h for famotidine and tizanidine loaded matrices respectively according to in vitro dissolution studies using USP type-II dissolution apparatus. Surface modification of BC matrices improved drug release retardant properties in pre-modification drug loading and discovered to be effective in controlling BC's drug release properties. As a result, these modified BC matrices have the potential to be used in modified drug delivery systems. Yang et al. (2018) developed a novel efficient adsorbent of bacterial cellulose/poly(*m*-phenylenediamine (BC/PmPD). The stable hybrid structure of PmPD nanoparticles functionalization on BC fibril was achieved after monomer (mPD) preadsorption on BC By in-situ oxidative polymerization of adsorbed mPD. The results showed optimum BC/PmPD has a Langmuir Cr(VI) adsorption capacity of 434.78 mg/g, which is significantly higher than that of many previously reported adsorbents. The study demonstrated that the BC/PmPD is a promising adsorbent for Cr(VI) removal due to its high adsorption capacity, efficient reclamation, and good regeneration performance.

Overall, it may be said the nanocellulose surfaces can be easily functionalized with several ionic or ionizable groups, which may improve the binding effectiveness towards various contaminants (Abouzeid et al., 2019). According to Abouzeid et al. (2019), the traditional method of TEMPO-mediated cellulose oxidation prior to mechanical defibrillation integrates negatively charged carboxylate groups on the surface of CNF, which can aid in the adsorption of heavy metal cationic species such as Ni^{2+} , Cr^{3+} , Cd^{2+} and Pb^{2+} . Furthermore, the surface hydrophilicity afforded by the TEMPO-oxidized CNF increased permeation flux and antifouling performance of the membrane for oily water separation (Huang, Yang, Yang, & Chang, 2021). Besides, TEMPO oxidation can specifically oxidise -OH to -COOH, enhancing methylene blue (MB) dye adsorption (Yang et al., 2017a, b). Thus, surface modification of nanocellulose by TEMPO-oxidation enhanced porous structure, adsorbed cations, and high surface area that could be used to adsorb dye and heavy metal ions from solution (Zhan et al., 2018).

6. Application of nanocellulose-based for wastewater treatment

Nanocellulose has been applied as a reinforcing agent in a wide range of polymer matrices, mainly water-soluble polymer matrices (Krause Bierhalz, 2021). The use of nanocellulose within the polymers can affect the tear strength, abrasion resistance, adhesion properties (Ilyas, Sapuan, Ishak, et al., 2019) and could acted as a pore forming agent (Arman Alim & Othaman, 2018). The specialties of nanocellulose for this particular application is due to their high aspect ratio and accessibility of plenty of –OH groups for the reinforcement with polymer composite (Ilyas, Sapuan, Atiqah, et al., 2020). Nanocelluloses have received much interest as a wastewater treatment material option (Salama et al., 2021) due to the availability of a large number of –OH



Fig. 6. Properties of nanocellulose as possible bio-sorbents for wastewater treatment (Reshmy et al., 2022).

groups in nanocellulose able to adsorb the dyes, heavy metals, and other contaminants in wastewater. Fig. 6 showed the properties of nanocellulose as possible bio-sorbents for wastewater treatment.

6.1. Nanocellulose-based adsorbents

Adsorption is a separation process which certain components of a fluid/gas phase are transferred to the surface of a solid adsorbent. Heavy metal ions, dyes, oils, and dissolved organic contaminants are among the chemical species that are often removed from wastewater using this approach. According to Köse et al. (2020), adsorption is the most popular and convenient method for removing non-degradable organic chemicals in low concentrations from drinking water or industrial effluents. It is because of cost-effective and efficient method to eliminate contaminants (both inorganic and organic). The nature of the pollutant, the quantity and type of the adsorbent, the presence of other contaminants, and the adsorption conditions (pH, contact time, particle size, temperature, initial concentration of pollutant, adsorbent dosage)are all factors that influence the adsorption process (Abouzeid et al., 2019).

Adsorbents are frequently applied in the form of spherical pellets, rods, mouldings, or monoliths with hydrodynamic diameters ranging from 0.5 to 10 mm, and must have good abrasion resistance, high heat stability, and tiny pore sizes, resulting in a larger exposed surface area. Fig. 7 showed the adsorbent materials that are often utilised in industry. Nanocellulose emerged as a great potential adsorbent material for the removal of pollutants from waste water due to its large surface area and plenty of hydroxyl groups to functionalize (Jaffar et al., 2022; Khulbe & Matsuura, 2018). However, surface functionalization is an important step for nanocellulose-based adsorbents in promoting pollutant adsorption and adsorption capacity (Mahfoudhi & Boufi, 2017). According to Mahfoudhi and Boufi (2017) the surface functionalization of nanocellulose can be accomplished by various surface modification procedures involving the hydroxyl function's chemistry (Fig. 5).

Xi et al. (2020) investigated the fabrication and arsenic removal performance of cellulose nanocrystal absorbents based on the "bridge joint" effect of iron ions via coprecipitation. According to the findings, the iron ions successfully connect the two dispersed polymers, inducing a large number of O—Fe—O bonds and providing more adsorption

active sites for the removal of seriously contaminated and high-toxicity As(III)/As(V). Arsenic adsorption was primarily chemisorption and monolayer adsorption. CNC-PEI-Fe(III) has the best As(III)/As(V) removal effect, with an adsorption capacity of 142.42/78.71 mg/g.

Chai et al. (2020) developed a simple method of producing novel pHsensitive nanoparticles from nanocellulose by cross-linking polyethyleneimine and glutaraldehyde. The biosorbent displayed fast adsorption during the first 10 min, and the As(V) adsorption capacity of the nanoparticles reached nearly 255.19 mg/g at pH 3, which was five times higher than the As(V) solution's initial pH (44.33 mg/g). Meantime, the adsorbent performed admirably even after eight regeneration cycles. This novel material has tremendous potential for removing arsenic contaminants and developing pH-sensitive materials. In 2018, Yang et al. conducted a study to fabricate a novel efficient adsorbent from bacterial cellulose/poly(m-phenylenediamine (BC/PmPD) for Cr (VI) removal. The results revealed that the maximum BC/PmPD has a Langmuir Cr(VI) adsorption capacity of 434.78 mg/g. This study demonstrated that the BC/PmPD is a promising adsorbent for Cr(VI) removal because of its high adsorption capacity, efficient reclamation, and good regeneration performance.

6.1.1. Adsorption of heavy metal

Heavy metal contamination has received much interest due to its toxicity, difficulty in decomposition, and build-up in living organisms. Because the other procedures have intrinsic limitations, including the generation of large amounts of sludge, lack of efficient, reactive operating conditions, and costly disposal, adsorption is the most efficient (Renu, Agarwal, & Singh, 2017). The exploration of nanocellulose-based adsorbents for heavy metal removal has sparked a flurry of research to date. Abou-Zeid et al. (2021) conducted experiment on the removal of Cu(II), Pb(II), Mg(II), and Fe(II) by adsorption process onto alginate/ nanocellulose beads. Alginate was combined with three types of nanocellulose (CNC, CNF and tri-carboxylate cellulose nanofibers, TPC-CNF) to eliminate the metal cations. The metal cations Cu^{2+} , Pb^{2+} , Mg^{2+} , and Fe²⁺ were efficiently removed by the modified bio-polymeric beads (Alg-CNF, Alg-CNC, and Alg-TPC-CNF). Alsaiari et al. (2021) conducted experiment to synthesis, characterized, and applied the polypyrrole functionalized nanocellulose for the removal of Cr(VI) from aqueous



Fig. 7. Adsorbent materials that are often utilised (Mahfoudhi & Boufi, 2017).

solution. The maximum adsorption capacity of 560 mg/g according to the Langmuir isotherm exhibited better performance for Cr(VI) absorption compared to previous. The results showed functionalized nanocellulose improved the adsorbent's effectiveness of Cr(VI) in two ways: adsorption and reduction. According to Qin et al. (2019), the modified CNFs record high equilibrium Cu²⁺ removal capacity of 115.3 mg/g at pH 5.0.

The possibility of using carboxymethyl nanocellulose (CMNc) stabilised nZVI for effective Cr(VI) reduction in aqueous medium has been examined (Kumar, Kardam, Rajawat, Jain, & Suman., 2019). With an initial concentration of 6 mg/L and only 0.015 g of the produced material, the findings imply that adsorption effectiveness is about 100%. Pb (II), Cu(II), Zn (II), Cd(II), and Mn(II) each had maximum adsorption capacities of 571 mg/g, 462 mg/g, 361 mg/g, 263 mg/g, and 208 mg/g, respectively, for the CNFs adsorbent (Mo et al., 2021). The adsorbent was capable of quickly removing Pb(II) species (87% and 100% of its equilibrium uptake in 2 min and 10 min, respectively). The ability of hydrogel composites based on chitosan-g-poly(acrylic acid) matrices filled with cellulose nanowhiskers (CNWs) to adsorb Pb(II) and Cu(II) ions from water was investigated in the Rodrigues, Carlos, Medina, and Fajardo (2019) study. Within 30 min, at pH 4.0, using 20 mg of the hydrogel composite containing 10% w/w of CNWs, the highest adsorption of Pb (II) (818.4 mg/g) and Cu (II) (325.5 mg/g) was obtained.

Silva et al. (2020) synthesized dual nanofibrillar-based bio-sorbent films made up of cellulose nanofibrils (CNFs) and lysozyme nanofibrils (LNFs) for the removal of mercury (II) from aqueous solutions. With a Young's modulus of more than 5 GPa and thermal stability up to 250 °C, this material has excellent mechanical properties. The Hg (II) removal effectiveness in ultra-pure and natural spring waters polluted with ambient realistic amounts of mercury is 50 g/L. The removal efficiency of Hg (II) is pH-dependent, reaching at 98% after 24 h. Wei et al. (2019) developed a magnetic hybrid aerogel by combining nanocellulose and ferroferric oxide (Fe₃O₄) nanoparticles for efficient heavy metal ion adsorption from water. The adsorption effectiveness of the hybrid aerogel on the Cr(VI) ion achieves the greatest value of 2.2 mg/g. Furthermore, the hybrid aerogel exhibits similar adsorption behaviour on plumbum (Pb)(II) and copper (Cu)(II) ions.

The incorporation of NFC to the vinyl imidazole (VIM) monomer reported by Zhang, Zhong, Xiang, Zhang, and Feng (2021) in the development of poly(VIM)/MFC cryogel composite for continuous and efficient heavy metal removal. Hysteresis loops changed slightly and energy loss coefficients were less than 23% during loading-unloading 200 cycles, highlighting the excellent shape recovery and fatigue resistance. Batch adsorption revealed that the poly(VIM)/MFC cryogel adsorption capabilities to Cu(II), Pb(II), Zn(II), Cd(II), Ni(II), and Co(II) were 87.4, 53.9, 48.6, 44.3, 23.8, and 20.1 mg/g, respectively. Xu et al. (2021) used one-step pyrolysis to successfully manufacture nanocellulose and thiourea, which could efficiently activate peroxymonosulfate (PMS) to breakdown sulfamethoxazole (SMX) in water. Furthermore, the effectiveness of SMX removal by this oxidation system was 2.3–3.1 times that of previous systems triggered by common metal oxides (such as Fe₃O, Fe₂O₃, and MnO₂).

6.1.2. Separation of Oil-water

Oil does not dissolve in water and instead creates a thick sludge. As a result, many solutions to the oil-water separation process were established. According to the functionality of sorbent materials, lignocellulosic material is a suitable choice for oil-water separation operations. Cellulose-based materials may be functionalized and changed in a variety of ways to change their characteristics, such as porosity and hydrophobicity (oleophilicity) (Doshi, Sillanpää, & Kalliola, 2018). The porosity of the materials is required for their usage as adsorbers, allowing oil spillages to be incorporated into their matrix (Fürtauer et al., 2021).

Ji et al. (2020) studied the biomimetic co-deposition of polyphenol-

substance tannic acid induced the grafting of renewable cardanolderived siloxane on a cellulose nanofiber (CNF) framework to form an eco-friendly and low-cost microporous aerogel to capture oil contamination. Low density and good hydrophobicity were critical for very effective oil droplet absorption. With maximal absorption capabilities of up to 108 g/g, the modified aerogel efficiently gathered different oils and organic solvents from water (chloroform). The magnetically induced selective oil absorption of superhydrophobic cellulose nanofibril/silica fibre/Fe₃O₄ nanocomposite aerogel was explored (Mi et al., 2020). The created nanocellulose composite aerogel has a high absorption capacity (weight gain of 3420–5837%), 100% separation efficiency, and a water contact angle of 150°.

A new aerogel was developed by adding nano bentonite into the dialdehyde nanocellulose and carboxymethyl chitosan mesh of a hydrogel precursor. This lightweight aerogel demonstrated maximal dye removal capacity of up to 29.842 g/g and 20.927 g/g during the first 5 min of the reaction, throughout a wide pH range for Bromophenol blue and Direct Blue 6, respectively, and up to 50 times its own weight in oil and organic solvents (Sharma et al., 2020). Zhou et al. (2021) conducted experiment of solvent-free approach nanocomposite aerogels of from nanocellulose and nano-alumina (NC/Al₂O₃), The results show that the NC/Al₂O₃ aerogel with a low density of 5.1 mg cm³ could achieve the best pore microstructures and the highest oil and organic solvent adsorption capacities when prepared with a nanocellulose in aqueous solution.

6.1.3. Extraction of dye

Dye contamination of water sources can cause serious environmental and health issues. Prolonged contact to the dye causes respiratory issues as well as skin irritation. Many of the dyes, in fact, are mutagenic and carcinogenic, increasing the risk of cancer. Meanwhile, dyecontaminated water is difficult to cure due to the colours' nonbiodegradable behaviour and resistance to oxidising chemicals, light, and heat adsorption is a well-known method used in academics and industry to remove colours from solutions. The adsorption of dyes on adsorbents is a simple and cost-effective approach that is commonly employed for big and complex processes (Muthu & Khadir, 2022). Many studies investigated the potential of functionalized nanocellulose as a dye sorbent. The applicability of a cellulose nanofibril-based aerogel generated from sago pith waste to the removal of methylene blue was investigated. The SPCNF aerogel, with a density of 2.1 mg/cm³, was effective in removing methylene blue (MB), with a maximum MB adsorption of 222.2 mg/g at 20 °C. The SPCNF aerogel demonstrated exceptional MB removal efficiency, with 5 mg and 20 mg of SPCNF capable of removing more than 90% and almost 99% of MB, respectively (Beh et al., 2020).

Li, He, Chen, and Zhao (2021b) established a green manufacturing of porous microspheres incorporating cellulose nanocrystal/MnO₂ nanohybrid for effective dye removal. Simple method for manufacturing porous microspheres based on CNC/MnO₂ by freeze-drying the airbubble templated emulsion in which sodium alginate (SA) was utilised as the crosslinked matrix. The color removal ratio of methylene blue (800 mg/mL) could be up to 95.4% in 10 min, and the equilibrium decolourization could reach 114.5 mg/g. Another study on methylene Blue removal from an aqueous environment using cellulose nanofibers (CNFs) derived from the petioles of the nipa palm tree and graphene oxide (GO) conducted by Nguyen and colleagues (Nguyen et al., 2022). Aerogel is stable in the aquatic environment and may be reused five times with an adsorption capacity of more than 90%. Furthermore, the aerogel exhibited a fast adsorption rate for methylene blue (MB) adsorption, absorbing more than 99% of water in less than 20 min.

Tan, Bing, Reza, Abdullah, and Horri (2018) developed nanocrystalline cellulose (NCC) flakes for adsorption-based methylene blue (MB) removal. NCC flakes have a high adsorption capacity (188.7 mg/g fixed at 0.7 g/L adsorbent dose, 25 $^{\circ}$ C and pH 6). Unlike NCC powder, NCC flakes were shown to be easily removed from MB-containing effluent. Further adsorption tests on NCC flakes revealed that 0.7 g/L was the optimal adsorbent dose, which aligned well with the Langmuir Isotherm. The average free energy value calculated from the Dubinin-Radushkevich isotherm was less than 8 kJ/mol while at various temperatures, the values ranged from -20 kJ/mol to 0 kJ/mol. Reusable and environmentally friendly biopolymer composite films based on poly (vinyl alcohol)/chitin/nanocellulose were prepared and described for methylene blue (MB) dye removal (Mok et al., 2020). Maleic acid (MA) was employed as a biopolymer composite film crosslinker. The highest adsorption capacity was 467.5 mg/g, and the PVA/CT10/NCC/MA30 composite has a high adsorption reusability, with an adsorption percentage of 83.67 \pm 1.08% at the fifth cycle.

The isolation of cellulose nanocrystal (CNC) from Carex Meyeriana Kunth (CMK) using a TEMPO oxidation and mechanical homogenization process for methylene blue (MB) removal from aqueous solution was conducted by Yang et al. (2017a, b). The results revealed that increasing the CNC dose increased the percentage of MB removed by CNC. The adsorption kinetics of produced CNC followed the pseudo second-order model, and the adsorption isotherms conformed well to the Langmuir model, with a predicted maximum adsorptive capacity of 217.4 mg/g, which was more than that of CNC extracted by acid hydrolysis Yang et al. (2017a, b). Vilela, Moreirinha, Almeida, Silvestre, and Freire (2019) synthesized zwitterionic nanocomposite membranes out of crosslinked poly(2-methacryloyloxyethyl phosphorylcholine) (PMPC) and bacterial nanocellulose (BNC) and evaluated them as water remediation techniques. The removal of two water-soluble model dyes, i.e. methylene blue (MB, cationic) and methyl orange (MO, anionic), from water was evaluated, and the results showed that both dyes were successfully removed under the studied conditions, with a maximum of ionic dye adsorption of approximately 4.4-4.5 mg/g.

A newly established approach based on periodate oxidation of Cladophora nanocellulose resulted in micrometer-sized 2,3-dialdehyde cellulose (DAC) beads for Congo red adsorption (Ruan, Strømme, & Lindh, 2018). The quick and high adsorption capacity of DACeCS beads for Congo red at pH 2 and good desorption characteristics at pH 12 suggested potential uses as a suitable material in chemical waste adsorption (Ruan et al., 2018). For crystal violet removal, Sorriaux et al. (2021) developed a bio-based and robust polydopamine coated nanocellulose/amyloid composite aerogel. In particular, the bio-based aerogel displayed high adsorption efficiencies of 93.1% in 30 min.

6.2. Nanocellulose-based membrane

Membrane technology for contaminant removal are gaining attention because these alternative methods provide more efficient treatment methods, require minimal energy and do not require the addition of chemicals into the waste system (Srivastava, Arthanareeswaran, Anantharaman, & Starov, 2011). According to Abouzeid et al. (2019), water purification membranes or filter technology are worthy technologies for water filtering due to their great efficiency and no secondary pollutants. Flux, transmembrane pressure, permeability and recovery are considered design parameters for membranes treatment plant (Abouzeid et al., 2019). Besides, exploration of materials and processes utilised in the fabrication of optimal membranes for waste water separation, such as self-supporting, thermally and mechanically robust, homogenous porous structures and anti-fouling behaviour, is significant. Therefore, nanocellulose with these features offer tremendous promise in membrane filtration applications and environmental remediation (Jaffar et al., 2022).

According to Jaffar et al. (2022), nanocellulose can be utilised as a matrix or an addition to increase membrane performance in water filtration as well as nano-dimensional properties related to high specific surface area, high aspect ratio, tunable porosity, outstanding reinforcing potential, high degree of crystallinity, tunable surface chemistry, and anti-fouling properties. Fig. 8 showed potential mechanism of



Fig. 8. Mechanism of nanocellulose membrane in wastewater purification (Reshmy et al., 2021).

nanocellulose membrane in wastewater purification. In addition, two uses of nanocellulose have gotten a lot of attention and have shown to be effective: as an adsorbent and as a membrane for pollutant removal (Abouzeid et al., 2019). According to Dufresne (2019), this potential is linked to its high aspect ratio, high specific surface area, excellent capacity retention, and environmental inertness. The existence of active sites allows for the integration of chemical moieties that may improve the effectiveness of pollutants adhering to the surface. Their nanostructure might be utilised to develop filters that clean a wide range of liquids. Aside from the aforementioned benefits, nanocellulose-based membranes are easy-to-fabricate and low cost (Yuan et al., 2020).

Cheng et al. (2019) developed a bacterial cellulose membrane (BCM) modified with ethylenediaminetetraacetic acid (EDTA) and crosslinked with (3-aminopropyl) triethoxysilane (APTES) to remove Sr(II) from water. The adsorption of Sr²⁺ followed the pseudo second-order kinetic model ($R_2 = 0.999$) and fit well with the Langmuir isotherm model (R_2 = 0.996). The maximum adsorption capacity was calculated to be 44.86 mg/g, which was comparable to the capacity of other adsorbents. Ferreira-Neto et al. (2020) investigate the fabrication of bacterial nanocellulose/MoS₂ hybrid aerogels as bifunctional adsorbent/photocatalyst membranes for in-flow water decontamination. The prepared BC/MoS₂ aerogel membranes demonstrated high performance in the photoassisted in-flow removal of both organic dye (MB) molecules (96% removal within 120 min, $K_{obs.} = 0.0267 \text{ min}^{-1}$) and heavy metal (Cr (VI)) ions (88% removal within 120 min, $K_{obs.} = 0.0012 \text{ min}^{-1}$) under UV-visible light illumination, as well as superb recyclability and photostability.

6.2.1. Heavy metal removal

Heavy metals are dumped into water from numerous sectors, and they can be poisonous or carcinogenic in nature, posing serious hazards for humans and marine ecosystems (Renu et al., 2017). Nanostructured membranes have recently been developed for the removal/rejection of pollutants based on physical/chemical properties (Karim, Mathew, Kokol, Wei, & Grahn, 2016; Khulbe & Matsuura, 2018; Mohd Noor et al., 2019). In 2016, Palliparambil et al. (2016) generate cellulose acetate membranes with and without zero valent iron nanoparticles (ZVI) for nickel removal from aqueous solutions by phase inversion approach. The removal of nickel ions suggests that membranes containing ZVI performed better. Continuously, membrane containing ZVI were conducted with the effect of variables such as contact time, initial pH, adsorbent dosage, and initial ion concentration on membrane adsorption capacity. The maximum nickel removal effectiveness was discovered to be 96%. Meanwhile, the addition of cellulose nanocrystals (CNCs-M) into nanocomposite polyethersulfone (PES) membranes increased porosity, hydrophilicity, elasticity, and pure water flow more than carbon nanotubes (CNTs-M) neat, reported by Bai et al. (2017). The results showed that nanocellulose is a viable alternative to carbon nanotubes (CNTs) due to its widespread availability, environmental friendliness, biocompatibility, biodegradability, renewable nature, higher hydrophilicity, strong mechanical qualities, and low cost.

Bai et al. (2020) expanded their work to develop cellulose nanocrystal-blended polyethersulfone (PES) membranes for improved natural organic matter (NOM) removal and minimization of membrane fouling. Surface morphology analysis found the CNC-containing nanocomposite membranes had homogenous surfaces. The results showed nanocomposite membranes have greater porosity, zeta potentials and pure water flux than the conventional PES membrane. The incorporation of CNC to PES membranes demonstrated improved antiNOM fouling capabilities, greater cleaning performance, and effective fouling control.

According to Karim et al. (2016), interactions between negatively charged nanocellulose and positively charged of metal ions are expected to provide the main mechanism for metal ion elimination. The removal of $(Ag^+ \text{ and } Cu^{2+}/Fe^{3+}/Fe^{2+})$ in mirror industry effluent was accomplished by employing cellulose microfiber sludge as a support layer and cellulose nanocrystals (CNCSL, CNCBE, or PCNCSL) in a gelatin matrix

as the functional layer. The results revealed that PCNCSL had a high ion removal capability, with a capacity of 100%, followed by CNCBE and then CNCSL.

6.2.2. Oil-water filtration process

Filtration is a viable choice among water treatment technologies for treating ground water, surface water, and waste water. Membrane technology has been identified as the most promising water filtering approach since it produces high-quality treated water without the use of chemicals while requiring little energy. The membrane functions as a particular filter, allowing water to pass through while retaining suspended particles and other impurities. Membrane filtration can be employed as an alternative to flocculation, sediment purification, adsorption, extraction, and distillation. A membrane filtering process's efficiency was assessed by its selectivity and productivity (membrane dependant) (Gupta & Ali, 2013). Subsequently, nanocellulose could be considered as a smart material for membrane filtration since it meets all of the requirements by delivering effective and cheap treatment procedures at a low cost using abundant renewable resources (Choudhury et al., 2020).

Cheng et al. (2017) conducted experiment on the fabrication of superhydrophilic membranes made of fibrous tunicate cellulose nanocrystals (TCNC) by facile-vacuum assisted method for highly effective oil/water separation. The results revealed that the TCNC membranes were advantageous for very efficient separation of oily water, capable of separating both oil-in-water and water-in-oil emulsions. The dosage of TCNCs might be used to adjust the thickness, pore size, water flux, and oil rejection of the TCNC membranes. They also have strong mechanical strength, good pH and temperature stability, and good cycle performance. In 2018, Zhan et al. investigate a simple method for fabricating durable membranes for multifunctional oil/water emulsion separation from renewable tunicate cellulose nanocrystal (TCNC) and low-cost palygorskite (PGS). The TCNC/PGS membranes have a nanoporous structure with adjustable thickness, as well as a superhydrophilic and underwater superoleophobic surface, allowing them to easily separate micro/nanoemulsions exhibiting high water flux and oil rejection. The membranes produced demonstrated high mechanical strength, outstanding recyclability, and good stability under demanding conditions. More crucially, TCNC/PGS membranes have the potential to remove water soluble pollutants (dye or heavy ions) during the oil/ water separation process, resulting in multifunctional water purification.

Meanwhile, tunicate cellulose nanocrystals (TCNCs) modified filter papers were fabricated by physical and chemical strategies, respectively conducted by Huang et al. (2019). TCNCs were directly coated on the surface of physically modified filter paper via hydrogen bonding, whereas TCNCs were fixed on the surface of filter paper for chemically by crosslinking hydroxyl groups with epichlorohydrin (ECH). The TCNC-modified filter sheets had a nanoporous shape and a super hydrophilic/underwater superoleophobic surface. Specifically, the resulting filter sheets were capable of efficiently separating diverse oil/water combinations and emulsions. Furthermore, these filter papers demonstrated excellent durability, retaining their inherent characteristics across a variety of acidic, basic, and salty environments, mechanical abrasion (or peeling), and ultraviolet radiation (365 nm).

Li et al. (2019) developed all-cellulose membranes by using a largescale additive printing approach to deposit cellulose nanocrystals (CNC) onto the surface of mixed cellulose esters (MCE). By adjusting the printing cycles of cellulose nanocrystal inks, the thickness, pore size, surface wettability, and water flux of all-cellulose membranes may be fine-tuned. The resulting all-cellulose membrane contains of nanoporous architecture (76–91 nm) exhibit superhydrophilicity and underwater superoleophobicity, allowing for the separation of oil/water nanoemulsions with a high-water flux (>1500 Lm⁻² h⁻¹ bar⁻¹) and ultrahigh efficiency (>99%) after optimal printing conditions. Additionally, these all-cellulose membranes have good stability and reusability for longterm separation, and their inherent features are retained through a variety of acidic, basic, and salty environments, as well as mechanical abrasion which is reported to have outstanding separation performance for oil/water nanoemulsions.

In 2021, Huang et al. conduct experiment to fabricated selfsupported nanoporous lysozyme/nanocellulose membranes for multifunctional wastewater purification (such as organic dyes, heavy metal ions, emulsified oil droplets, and toxic chemicals). Lysozyme oligomer was able to shrink the pore size of the resulting membranes to a precise cut-off size of 3 nm by acting as an adhesive to bond cellulose nanofibers generated via 2,2,6,6-tetra methyl-1-piperidinyloxy oxidation (TEMPO). The resulting nanocomposite membranes could effectively eliminate organic dyes, heavy metal ions, bilirubin, oil droplets, and boron from wastewater due to the numerous functional groups (–OH, –COOH, –SH, –NH₂). Interestingly, these membranes demonstrated outstanding recyclability, mechanical performance, and acid/alkali resistance presented a rapid, cost-effective, and long-term strategy for the manufacture of multifunction membranes for treating wastewater using natural resources and biodegradable nanomaterials.

Almeida et al. (2020) investigated the production of cellulose acetate (CA) electrospun non-woven membranes stamped with various cellulose nanocrystal (CNC) patterns as selective oil microdroplet removal from water emulsions with an effectiveness of up to 80%. Yin et al. (2020) developed a cellulose nanofiber (CNF) polyelectrolyte filtration membrane with superior underwater superoleophobicity and antifouling performance. The membrane has a high separation efficiency (>99%) in separating oil/water emulsions. The combination of the annealed cellulose nanoparticles with non-woven electrospun membranes opens the door to a low-cost environment friendly method to remove oil microdroplets from polluted ocean and waste-oily waters.

Wang et al. (2019) used top-down wood nanotechnology to create an anisotropic, durable, and superhydrophobic aerogel with a water-responsive shape-memory function for multi behavioural and reusable oil/water separation. For excellent oil/water separation, the filtering and absorption functions were organically integrated in the nano-cellulose wood aerogel/PDMS composite. Because of the quick water-responsive shape-memory functionalization, the as-prepared wood aerogel/PDMS bulk composites displayed substantial oil absorption (approximately 20 g/g) and reusability. The produced wood aerogel/PDMS membrane materials separated oil/water mixtures with excellent efficiency (99.5%) and flux (about $2.25 \times 104 \text{ L/m}^2 \text{ h}$).

Cruz-Tato et al. (2017) investigate the possibility of fabricating the support layer of thin film composite (TFC) membranes for water purification applications utilising nanocellulose (NC)-based composites with silver and platinum nanoparticles as additive materials. The membranes were created with finger-like pore morphologies and variable pore diameters. With wastewater samples, the MNC-TFC membranes produced greater water fluxes and solute rejection. For brackish water desalination, nanocellulose acetate nanofiltration (CA-NF) membranes were manufactured via a phase inversion procedure. A greater performance was obtained using a composite membrane manufactured at a rate of 20% and at a temperature of 90 °C. According to the study, utilising reinforced PTFE improved the composite membrane's performance in terms of water permeability, flux, and salt rejection by 20% over standard CA-membranes (Shaaban, El-Khateeb, & Saad, 2019).

Then, Ma, Burger, Hsiao, and Chu (2014) fabricated CNF membranes with varying degrees of oxidation as a function of sodium hypochlorite content (10–15 wt%). Other factors varied were CNF loading (0.01–0.20 wt%), ionic strength (0–0.083 mol/L NaCl), and pH (4 and 10). The shape of the membrane, as well as its thermal stability and durability, were all assessed. When compared to PAN10 membrane, CNF-based membrane demonstrated a 5-fold improvement in permeate flow while achieving equivalent microsphere rejection (>99.7%). In the oil-water separation, the CNF-based membrane demonstrated good rejection of oil (>99.5%) with permeate flow, which was about 8 times more than the permeate flux of the PAN10 membrane.

6.2.3. Dye separation

Water pollution is a serious global problem caused by the direct dumping of toxic dyes into the environment. When dyes are present in water, they can be hazardous to aquatic species and people. Dyes are naturally recalcitrant, with the ability to withstand attacks from heat, light, and bacteria. As a result, the majority of reactive dyes are nonbiodegradable, and their removal from aqueous solutions is extremely difficult. There are several approaches for treating dye-contaminated water, with membrane technology emerging as one of the most promising. Membrane separation has gained popularity due to key benefits such as simplicity of implementation, cheap cost, and waste reduction (Muthu & Khadir, 2022).

Karim, Mathew, Grahn, Mouzon, and Oksman (2014) fabricated pure biobased composite membranes for water purification by freeze-drying and compacting cellulose nanocrystals (CNCs) in chitosan matrix for water purification. The CNCs were bonded in a stable and nano-porous membrane structure with a thickness of 250–270 μ m by chitosan (10 wt %), which was further stabilised by cross-linking with gluteraldehyde vapours. Despite the modest water flux (64 L m²/h), the membranes efficiently eliminated 98%, 84%, and 70% of positively charged dyes such as Victoria Blue 2B, Methyl Violet 2B, and Rhodamine 6G during a 24-h contact period.

Liu, Zhu, and Mathew (2019) used two successive vacuum filtering stages to coat an ultrathin Graphene Oxide (GO) layer on a CNF membrane, resulting in negative charge content for dye solution filtration. At low GO concentration, the standing GO/CNF membrane surface was found, and they produced multiple mass transport nanochannels to achieve the outperformed water flux of $18,123 \times 574 \text{ Lm}^2/\text{h}$ bar. The combined effects of adsorption and size exclusion lead to the retention of three dyes (Victoria Blue 2B, Methyl Violet 2B, and Rhodamine 6 G) of more than 90% at 0.9 bar. In another study, Yin et al. (2020) developed a Cellulose nanofiber (CNF) polyelectrolyte filtration membrane with superior underwater superoleophobicity and antifouling performance for dye removal. The membrane removes positively charged dyes, including a series of methylene blue (MB) and gentian violet (GV), with high penetration fluxes (>10,000 L m²/h) and rejection ratios (>98%).

On the other hand, nanocellulose-based membranes can be used for a range of applications other than adsorption and water filtrations, such as gas separation, photocatalysis, pollution sensors, and energy devices. Another important application of nanocellulose-based membranes is gas separation. Membranes allow CO_2 to pass from the feed side to the permeate side in a selective manner (Jaffar et al., 2022). In 2017 Hosakun et al. (2017) used bacterial cellulose (BC) as an active material for membrane fabrication of CO_2 capture. The results showed that CO_2 has a strong interaction with the BC membrane, which is further enhanced by its modification experiment, the basic BC membrane, silk fibroin-modified BC membrane, and ZnO nanoparticles-modified BC membrane had CO_2 permeabilities of 2.73, 2.69, and 2.66 Barrer, respectively.

In addition, the nanocellulose-based membrane could be use in the sensor applications. The recent developments of nanocellulose-based sensors including electrochemical, optical, colorimetric, fluorescent, and bio-sensors for the detection of various types of pollutants such as heavy metal ions, water-soluble gases, minerals and salts summarized by Jaffar et al. (2022) and Fang, Hou, Chen, and Hu (2019). According to Jaffar et al. (2022), nanocellulose contains a number of hydroxyl groups that can be used to accommodate binding sites for selective analyte species adsorption, increasing electrochemical sensor selectivity, sensitivity, and durability. The presence of a large number of carboxyl and hydroxyl groups caused the sensors' ionic conductivity to adjust and adapt to gas exposure. Furthermore, nanocellulose-based materials could offer great potential in the development of renewable energy devices such as fuel cells, solar cells, and generators to tackle the real challenges of environmental sustainability. Ram, Velayutham, Sahu, Lele, and Shanmuganathan (2020) used polydopamine coated nanocellulose to develop a Nafion composite membrane. The addition of polydopamine-coated nanocellulose (PNC) improved Nafion's mechanical and thermomechanical stability up to 100 °C. PNC/Nafion membranes demonstrated significantly lower creep compliance (higher dimensional stability under constant stress) at 30 and 60 °C, which could be useful in the fabrication of membrane electrode assembly (MEA) as well as improved membrane durability in fuel cells.

7. Sustainability, limitations and challenges

Water pollution is thus mostly caused by the discharge of polluted wastewater containing dangerous compounds into surface or groundwater and primarily reliant on wastewater treatment. Adsorption is the most common and easy method for removing pollutants from wastewater due to it cost-effective and efficient way of removing pollutants (Reshmy et al., 2022). The usage of green materials derived from natural sources as bio-sorbents for wastewater pollutant removal is gaining such attention. Adsorbents made of nanomaterials have recently been used to enhance adsorption capacity, selectivity, and regeneration (Khulbe & Matsuura, 2018). When compared to other materials, cellulose's relatively large and ubiquitous characteristics to modify into derivatives such as nanocellulose make its fabrication cost-effective (Chen et al., 2019), give superior mechanical properties, allowing for greater surface area and pore volume (Reshmy et al., 2021), excellent biocompatibility and adaptable surface chemistry (Yuan et al., 2020). In addition, nanocellulose could be easily surface-modified to develop novel binding sites and give specialised features for adsorbing various types of contaminants.

However, significant modifications are necessary to overcome the restrictions associated with low thermal stability and incompatibility with a variety of nonpolar thermoplastics of nanocellulose which is costly, longer period and risks (Krause Bierhalz, 2021). The distribution of hydrophilic nanocellulose in hydrophobic polymer matrices is challenging and requires surface grafting with low molecular weight polymers (Jaffar et al., 2022). Additionally, nanoparticles cause a large pressure drop if packed in an adsorption column. Therefore, nanoparticles must either be immersed in or coated on the surface of larger particles, compromising their effectiveness (Khulbe & Matsuura, 2018). The use of huge amounts of adsorbents and additional chemicals to maintain a pH that allows for adsorption are the challenges in removing heavy metals from wastewater. (Renu et al., 2017). Besides, establishing cost-effective industrial procedures for nanocellulose synthesis and scaling is important for commercialization (Jaffar et al., 2022).

According to Khulbe and Matsuura (2018) the use of polymeric membranes for hazardous pollutant adsorption could potentially contribute to the evolution of next-generation reusable and portable water treatment systems. The dual function of membrane filtration and adsorption to be particularly successful in removing trace levels of contaminants such as cationic heavy metals, anionic phosphates, and nitrates (Khulbe & Matsuura, 2018). Interestingly, the benefits of nanocellulose-based filtration membrane include its high porosity and excellent hydrophilicity, good mechanical stability, great chemical inertness, and antifouling capability, making it useful in industrial wastewater treatment (Liu et al., 2021). Furthermore, nanocellulosebased membranes are simple to manufacture and inexpensive (Yuan et al., 2020).

However, one of this material's shortcomings is that isolating cellulose components is energy-intensive, and necessitates the use of hazardous chemicals that are harmful to humans and the environment (Phanthong et al., 2018; Tshikovhi et al., 2020). Furthermore, the development of technology to produce large-scale industrial manufacturing has been difficult (Yuan et al., 2020). To address this, attempts are being made to identify novel ways capable of enhancing the current process or fostering large-scale artificial synthesis. The high energy demand of nanocellulose modification, membrane manufacturing processes, the time-consuming, high cost regeneration of reusable membrane and the degradation of nanocellulose composite membranes may pose major challenges for scale-up applications of dual functional membranes that are adsorptive and separative (Reshmy et al., 2021; Yuan et al., 2020). Nanocellulose-based membrane filtration technology is still being developed to ensure high membrane flux, increase the rejection rate under different filtration conditions, reduce membrane pollution, and ensure service life of the filtration membrane (Liu et al., 2021).

8. Conclusions and perspectives

The application of adsorptive membranes to harmful contaminants could contribute to the development of effective and recyclable wastewater treatment for future generations due to the worldwide environmental issues. The removing of various contaminants in wastewater such as dyes, heavy metals, organic compounds and oils by nanocellulose-based adsorbent gained substantial attention because of its exceptional binding and adsorbing capacities which could be modified using a variety of approaches to increase their performance. It was found that optimal values of parameters such as the nature of the pollutants, the type of nanocellulose, and adsorption conditions (pH, contact time, particle size, temperature, initial concentration of pollutant, nanocellulose dosage) should be properly considered in the adsorption performance of contaminant removal. For filtration process, the modification and dosage of nanocellulose effected the thickness, pore size, water flux, and contaminants adsorption of the membranes. Moreover, the corporation of adhesive and additive materials with nanocellulosebased membrane demonstrated outstanding recyclability, mechanical performance, and acid/alkali resistance enhanced multifunction membranes for treating wastewater. The nanocellulose modification aided in the development of novel biomaterials as dual-functional membranes for the effective removal of contaminants in wastewater. This hybrid separation process improves the ability to remove pollutants quickly and efficiently. The understanding of nanocellulose isolation method, modification and properties as well as membrane adsorption was conducted for future work.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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