

Current Cellulose Nanofibrils and Cellulose Nanocrystals as Water Purification Functional Membrane Materials



ABSTRACT

This study highlights the beneficial role of cellulose nanofibrils (CNFs) and cellulose nanocrystals (CNCs) as components in functional membranes. The approaches of the use of CNF and CNC as membrane materials for water purification have been studied extensively during the past decades. This is due to their inherent abundance, renewability, sustainability and unique properties such as high aspect ratio, high surface area, high crystallinity, and high mechanical properties. The performance of CNF- and CNC-based membranes especially in treating actual water samples were also highlighted in this review to give a better overview of the behavior of these nanocellulose as membrane materials. The challenges of using CNFs and CNCs and the need for improvements for the future development of membrane materials are also discussed.

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INTRODUCTION

Inadequate access to clean water is one of the most pervasive problem humans are facing globally and with water scarcity becoming a serious threat, it is predicted to worsen in the future (Ying *et al.* 2017). World Health Organization (WHO) and the United Nations Children's Funds (UNICEF) estimated that 2.1 billion people still lack access to safe and readily available water at home, and about 800,000 people are killed every year as a result of consuming contaminated drinking water, inadequate handwashing facilities and inappropriate sanitation services (WHO and UNICEF 2017; Sogani and Vyas 2019). Therefore, development of efficient and sustainable water purification techniques is essential. Among the water purification techniques currently being developed, the membrane filtration approach is increasingly emerging today. This is due to its high separation selectivity, low energy consumption, no required additional chemical

additive and no regeneration of spent media (Pendergast and Hoek 2011; Ying *et al.* 2017). Membranes are defined as barriers that separate two phases and enable to restrict the transport of various components in selective manner (Wang *et al.* 2008). Membrane technology is available for solids, viruses, protein and bacteria removal, among others. Membrane filtrations are classified into microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) depending on the particle size being filtered and the membrane porosity (**Figure 1**).

Membranes are categorized based on structure and materials (**Table 1**) (Lee *et al.* 2016; Madhura *et al.* 2018). For the structure, membranes are grouped into two: isotropic membranes (homogenous in composition), which includes microporous membranes (Thakur and Voicu 2016; Zou and Zhu 2018), nonporous dense films

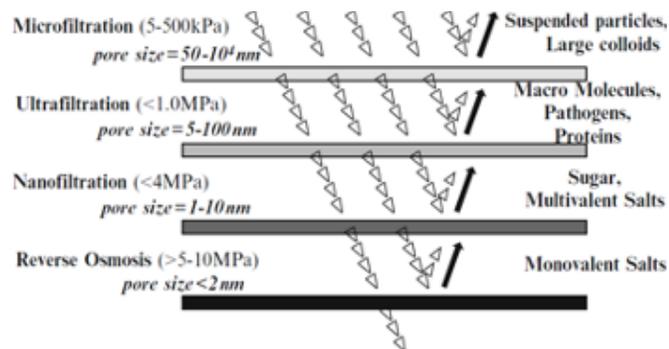


Figure 1. Classification of membrane filtration (Wang et al. 2008).

Table 1. The classification of membrane based on structure and materials.

Classification	Groups	Sub-Classification
Structure	Isotropic	Micro porous Membrane Non-porous Dense Film Membrane
	Anisotropic	Electrically-charged Membrane Phase-separation Membrane Composite Membrane
Material	Organic or polymeric materials Inorganic Hybrid of Organic and Inorganic	

(Al-saffar et al. 1997; Ong et al. 2016) and electrically charged membranes (Zydney 2015, Ahmed et al. 2016); and anisotropic membranes (heterogenous in composition), which includes phase-separation membranes (Mat Nawi et al. 2020) and composite membranes (Zahid et al. 2018; Mat Nawi et al. 2020) such as thin-film (Song et al. 2005; Rana et al. 2011; Lau et al. 2012), coated films (Fathizadeh et al. 2017; Ramanan et al. 2018; Fuwad et al. 2019), and self-assembled structures (Van Rijn et al. 2013; Bitton et al. 2014). Microporous membrane has a pore size about 0.1–5 μm and can only be used for filtering particles with 1–10 μm , which limits their applications in water purification (Lalia et al. 2013). Meanwhile, nanoporous membranes exhibited high performance for water purification as it can filter most of the pollutants (Gao et al. 2014).

Membranes can be made from organic (polymeric) materials, inorganic materials or a combination of the two. Organic membranes are those made of polypropylene, poly (vinylidene fluoride), polysulfone, poly

(ethersulfone), polyacrylonitrile, cross-linked aromatic polyamide and cellulose acetate (Grosso et al. 2014; Altintas et al. 2016; J. Miller et al. 2016; Ying et al. 2017; Bassyouni et al. 2019; Sapalidis 2020), while inorganic membranes are made mostly of ceramics, metals, metal oxides and carbon ceramics (e.g. Al_2O_3), metals (e.g. Cu, Ag, Au), metal oxides (e.g. TiO_2 , ZrO_2 , ZnO) and carbon based (e.g. graphene oxides, carbon nanotubes and zeolites) (Mohmood et al. 2013; Kumar et al. 2014; Castro-Muñoz 2020). The latest development in membrane material design is the use both organic and inorganic materials to produce a composite membrane.

Regardless of membrane type, the common goal is to have high flux, permeation, and high solute rejection; mechanical, chemical and thermal stability; cost-effective; and good chemical and fouling resistances (Sagle and Freeman 2004; Hwang et al. 2017). Large-scale applications of inorganic membranes are still limited due to high operation cost and inherent mechanical fragility despite having higher combinations of stability compared to polymeric membranes (Jin et al. 2010; Hofst et al. 2011). Thus, in water purification, the most widely used membrane types are organic or polymeric membranes (Bassyouni et al. 2019; Hadi et al. 2019). However, further commercialization of polymeric membrane is bound to several disadvantages such as more undesired expenses, high energy demand (Ladewig and Al-Shaeli 2017; Ying et al. 2017) and vulnerability to thermal, chemical and biological degradation (Wang et al. 2017). Furthermore, issues such as high hydrophobicity, low flux, membrane fouling, and low mechanical strength are yet to be solved. Remarkable progress have been made in decades in establishing new fabrication method and modification tailoring membrane pore structures, surface properties, and morphology (Wang et al. 2018; Aziz et al. 2020). These various fabrication methods being developed includes incorporation of nanomaterials in a polymer matrix. Cellulose nanomaterials, among other recent advances in nanotechnology, have been recognized to show great potential as functional membranes and enhance the performance of conventional membrane materials (Carpenter et al. 2015; Abouzeid et al. 2019).

Cellulose is the most abundant polymer, serving as a structural component in the cell wall of green plants (Heinze 2001; Mahfoudhi and Boufi 2017). Various source of cellulose includes plants, wood, bacteria, algae, fungi animals, (Trache et al. 2017) and waste paper (Kumar et al. 2020). It is characterized by its biocompatibility, hydrophilicity, good mechanical strength, stereo regularity and multichirality. The reactive hydroxyl groups in cellulose allow for the tuning

of desired properties through chemical derivatization (Heinze and Liebert 2001). Nanocellulose can be categorized into two groups; cellulose nanofibrils (CNF) and cellulose nanocrystals (CNC). CNF and CNC have been studied extensively during the past decade as membrane materials (Carpenter et al. 2015; Gopakumar et al. 2016); and elaborated below.

Recently, some reviews described detailed overview of the nanomaterial-based membrane development and its advances on water purification. Herein, this review is intended to present a brief overview of the different characteristics of and performance of nanocellulose (CNC and CNF) membranes in treatment of actual samples as well as the challenges encountered when using it as a membrane material for water purification.

MATERIALS AND METHODS

The study consisted of potentially relevant data from the year 2007 to 2020 of CNF- and CNC based membrane as the represent the recent published works more than 10 years. Data were extracted from each study and entered into standardized template. Selected studies were compared and summarized on the basis of the authors' perspective as well as existing theories and models. This study was carried out in January-June 2018 at KTH Royal Institute of Technology, Stockholm, Sweden.

RESULTS AND DISCUSSIONS

The subsequent sections elaborate the results of the review of current development of CNF and CNC as water purification functional membrane materials. The performance of CNF and CNC-based membranes (flux and adsorption capacities) is also highlighted in this review to give a better understanding of the behavior of these nanocellulose as membrane materials.

Cellulose Nanofibrils (CNF) and Cellulose Nanocrystals (CNC)

Cellulose is a polysaccharide composed of D-glucopyranose units linked by β -1,4-glycosidic bonds (Figure 2). The ability to form hydrogen bonds significantly influences the cellulose properties, structural function and its solubility (Jarvis 2003, Azizi Samir et al. 2005; Zhang et al. 2013; Rojas 2016). Cellulose has a large number of hydroxyl groups which provides extensive possibilities for intramolecular and intermolecular to form hydrogen bonds. Through these intermolecular hydrogen bonds and van der Waal forces, cellulose fibrils

are formed, which further aggregates into larger microfibrils (Moon et al. 2011, Salas 2014). Wood (hard wood and softwood), such as bleached birch chemical wood (Liimatainen et al. 2012), black spruce and eucalyptus (Fahma et al. 2010), are currently the most common sources of cellulose nanofibrils and are used for membrane fabrication (Stelte and Sanadi 2009). However, wheat straw soy (Alemdar and Sain 2008), bamboo (Chen et al. 2011), sugar bagasse (Teixeira et al. 2011), flax, hemp, abaca (Saba et al. 2014) and palm fruit stalk (Ghori et al. 2018) are also possible sources of cellulose. CNF or CNC are obtained from several processes such as mechanical, chemical or a combination of both or enzymatic disintegration of cellulose fibers. They only differ in the presence of unstructured regions (amorphous) and the length scale dimensions (Salas 2014) (Figure 2).

CNF, which contains both amorphous and crystalline domains, are 5-50 nm in width and within the microscale in length, (Mahfoudhi and Boufi 2017). CNFs are prepared by mechanical disintegration commonly using high pressure homogenizers (with range between 16-30 passes through refiner treatments) (Nakagaito and Yano 2004). Grinders (with 1-30 times passing through), microfluidizer (55–210 MPa with 5-10 passes), or cryocrushers (Zhang et al. 2013; Kargarzadeh 2017, Mahfoudhi and Boufi 2017) may be used to release elementary cellulose nanofibrils by breaking down the fiber cell wall (Figure 3). High energy consumption is required for mechanical disintegration, thus, preliminary 2,2,6,6-tetramethylpiperidine-1-oxyl radical (TEMPO)-mediated oxidation, carboxymethylation, mild acidic or enzymatic hydrolysis of cellulose, and certain other pretreatments are done to significantly decrease energy consumption (Kargarzadeh 2017). Pretreatments can be facilitated either through mild acidic or enzymatic hydrolysis of cellulose fibers and/ or through the introduction of charged groups onto cellulose chains, which can be accomplished during pulping or bleaching step or by subjecting cellulose fibers to oxidatives treatments, such as TEMPO-mediated oxidation (Isogai et al. 2011).

CNC, on the other hand, contains crystalline domains only. These are 5-70 nm in width (Carlmark et al. 2012) and have a length of 100-200 nm (Habibi et al. 2010; Brinchi et al. 2013; Xu et al. 2013). CNCs are also known as nanowhiskers, nanorods or rod-like cellulose crystals (Kargarzadeh 2017). These are mainly prepared by acid hydrolysis, typically using sulfuric acid or hydrochloric acid, of bleached cellulose fibers and fibrils, which degrades the amorphous region and releases highly crystalline nanoparticles (Zhang et al. 2013; Mahfoudhi and Boufi 2017).

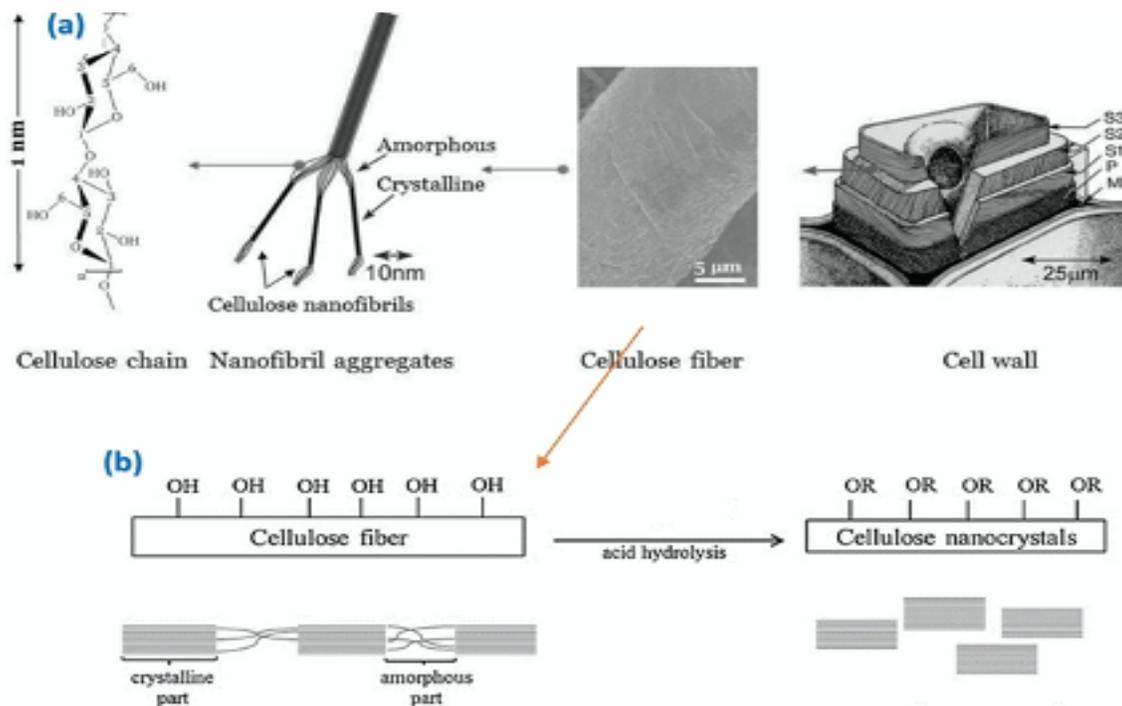


Figure 2. Schematic Illustration of (a) Cellulose nanofibril obtained from plant sources (Boujemaoui 2016) (b) Cellulose nanocrystals (Boujemaoui et al. 2015).

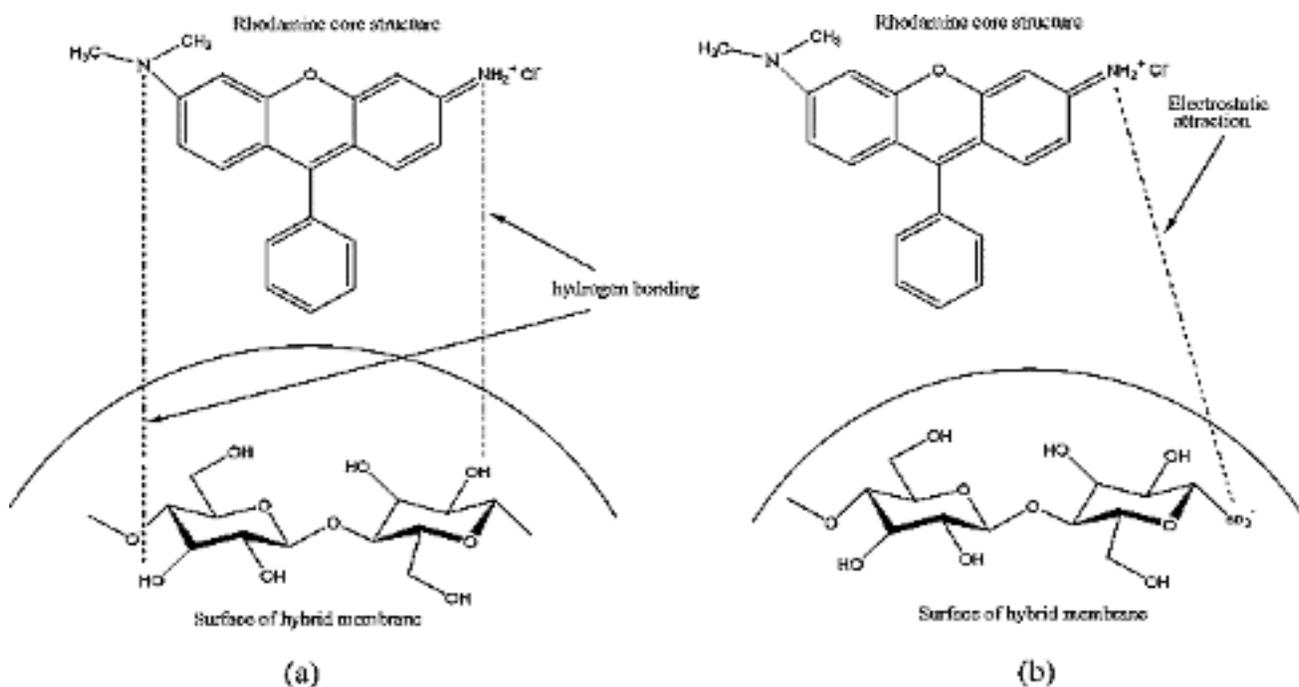


Figure 3. A schematic representation of the mechanisms of binding of the dyes with Cellulose Nanocrystals via (a) hydrogen bond formation and (b) electrostatic attraction (Karim et al. 2014).

CNF and CNC as Membrane Materials for Water Purification

The inherent abundance, renewability and sustainability, and unique physical and chemical properties of CNFs and CNCs have brought much

attention into the biopolymer world. These unique properties, such as high aspect ratio, high surface area, high crystallinity, transparency, and high mechanical properties, make CNFs and CNCs as important components in the membrane industry.

Characteristics of CNF and CNC

Cellulose nanofibrils and cellulose nanocrystals' inherent fibrous nature and their remarkable mechanical properties, having a tensile strength of 2-6 GPa (Carpenter et al. 2015) and elastic modulus between 130 GPa and 250 GPa (Lavoine et al. 2012), define their huge potential as components in water purification membrane. The hydrogen bonding makes the cellulose mechanically stable as it allows the chains to group together in a highly ordered structure which provides stiffness to cellulose chains (Suhas et al. 2016). Good mechanical strength offers resistance in high-pressure environment in real water purification applications. Moreover, the surface of nanocellulose can easily be modified through functionalization of the hydroxyl groups which increases the binding efficiency of pollutants to nanocellulosic materials. One of the most common protocols for the preparation of CNF is to pre-treat the pulp using TEMPO-oxidation as it will facilitate the defibrillation of the fibers yielding high concentrations of surface hydroxyl and carboxylic acid groups that can be further used during its modification or membrane fabrication. Incorporating CNF and CNC

as a reinforcement/ functional entity in various synthetic and natural polymer matrices such as chitosan (Karim et al. 2014), poly(acrylonitrile) (PAN) (Yang et al. 2014), poly(ether sulfone) (PES) (Daraei et al. 2016), poly(vinyl alcohol) (PVA) (Wang et al. 2013) and cellulose acetate (Battirola et al. 2017) or as hydrophilic modifiers (Kong 2014) has also received much attention in the membrane industry due to improved properties (Table 2). The different properties of CNF and CNC potential as membrane for water purification, were summarized and expanded in this section (Table 3).

Hydrophilicity. Fouling, the unwanted deposition and growth of microorganisms on surfaces caused by living organisms/non-living substances that result in the change of the surface properties, has shown severe impact on the performance of membrane in terms of flux and the lifespan (Mansouri 2010; Kong et al. 2014). Most of these problems are caused by the hydrophobic character of the membrane material. Thus, hydrophilic modifiers such as CNF have been employed to different polymeric membrane materials to reduce this problem (Uzal et al. 2017). However, it was observed that TEMPO-CNFs carrying carboxylic groups exhibit even

Table 2. Overview of the performance of 2,2,6,6-tetramethylpiperidine-1-oxyl radical-oxidized Cellulose Nanofibrils-based membranes with respect to different adsorption of heavy metals and percent rejection of different contaminants.

Contaminants	pH	Q_{max} (mg g ⁻¹) Max. adsorption	Rejection	Reference
Cr (VI)	4.0	87.5	-	Yang et al. 2014
	4.0	100		Wang et al. 2013
Pb (II)	5.0	137.7	-	Yang et al. 2014
	5.0	260	-	Wang et al. 2013
Ag (I)	-	0.87	-	Karim et al. 2017
Cu (II)	-	374	-	Karim et al. 2017
Fe (II)	-	456	-	Karim et al. 2017
Protein (BSA, EA, pepsin and trypsin)	-	-	50.5 %	Kong et al. 2014
Oil	-	-	99.5 %	Cao et al. 2013
E. coli	-	-	100 %	Wang et al. 2013
MS2 virus	-	-	LRV of 4	Wang et al. 2013

Table 3. Properties of Cellulose Nanofibrils and Cellulose Nanocrystals as membrane materials for water purification.

Property of CNF and CNC	Positive Effect on Membrane for Water Purification	Reference
Hydrophilicity Higher surface area	Reduces membrane fouling Better adsorption capacity	L. Kong et al. 2014 R. Wang et al. 2013; Carpenter et al. 2015; Sato et al. 2011
Porous Broad possibility of surface functionalization High mechanical stiffness Reusability	Enhance permeability and rejection efficiency Greater active sites for interactions of pollutants Increase the strength of the membrane High regeneration efficiency	Visangko et al. 2014 Yang et al. 2014 Karim 2014 R. Wang et al. 2013; Yang et al. 2014
Antibacterial High crystalline degree	Enhance bacterial resistance Enhance chemical resistance	Hassan et al., 2017 Ma et al. 2014

higher hydrophilicity, thus providing improved anti-fouling properties (Mautner 2020). The study of Kong *et al.* (2014) showed that the increase of the surface hydrophilicity of cellulose triacetate ultrafiltration (CTA) membranes through the addition of TEMPO-oxidized cellulose nanofibrils (TOCN) provided a higher recovery ratio compared to the CTA membrane alone. Higher recovery ratio means better antifouling property and higher efficiency of the hydraulic cleaning as protein molecules absorbed by the TOCN/CTA UF membrane can be more easily washed away (Kong *et al.* 2014). In another study, Hadi *et al.* (2019) prepared a thin-film nanofibrous composite (TFNC) membrane with electrospun mat as substrate and hydrophilic nanocellulose as the antifouling barrier layer. It resulted to a lower fouling tendency (<10%) and a higher degree of protein rejection ratio compared with the conventional membranes (fouling tendency >30%) .

Similar to the carboxylic groups in TEMPO-CNFs, the high surface charge provided by sulfate half esters of CNC is responsible for reducing the fouling of membranes. Ma *et al.* (2012) fabricated a nanofibrous microfiltration (MF) membrane by an electrospun polyacrylonitrile (PAN) nanofibrous scaffold supported by PET non-woven substrate infused with ultrafine CNC and compared its performance with GS0.22, a commercial membrane. Based on their results, GS0.22 showed a dramatic decrease in permeation flux due to fouling of tested particles due to the hydrophobic interaction. However, the fouling was reduced by the addition of CNC due its surface hydrophilicity (Ma *et al.* 2012). The CNC incorporated with hydrophobic poly(vinylidene fluoride) (PVDF) membrane via phase inversion improved the permeance flux (about 20 times) compared to the pristine PVDF membrane and the fouling tendency was decreased by 48.8% (Lv *et al.* 2018).

Higher Surface Area. The large surface area of CNF and CNC provides benefits such as better adsorptive removal. This material property provides more efficient adsorption capability thru charge interactions. Wang *et al.* (2013) showed complete removal of *E. coli* by size exclusion through a MF membrane composed of a two layered nanoscale PAN/PET fibrous scaffold with ultrafine CNF while maintaining good permeation flux (Wang *et al.* 2013). This material characteristic was also shown in the study of Sato *et al.* (2011) where the material completely removed bacteria using a nanofibrous composite membrane with an ultrafine CNF network due to its high surface-to-volume ratio (Sato *et al.* 2011). Moreover, Ma *et al.* (2012) found that the maximum adsorption capacity of MF membranes with ultrafine CNC for crystal violet dye

were 16 times higher than the commercial membrane.

Porosity. Membranes are characterized by porosity. The more porous it can be, the higher selectivity and separation the membrane can have (Geise *et al.* 2010). Addition of CNF or CNC into polymeric membranes improve the porosity which enhance the permeability, and the rejection efficiency. Additionally, membranes are characterized by their molecular weight cut-off (MWCO). This is a method that describes the pore size distribution and retention capabilities of the membrane, i.e. the lowest molecular weight of solute with a rejection of ninety percent (Li *et al.* 2017).

Thickness greatly affects the porosity of the membrane and inversely correlated with permeation flux. Flux is dependent on the water characteristics, applied pressure and on membrane properties such as pore size, surface porosity and pore density (Van der Bruggen *et al.* 2003). As fluid passes through a membrane, pores become blocked with particles and hence reducing the pore density, resulting in a drop in the filtration rate. This was shown in the study of Kong *et al.* (2014) wherein addition of TOCN to CTA (cellulose triacetate) membrane lead to an increase of water flux from 90.57 to 224.68 L hr⁻¹ m⁻². However, increasing the percentage weight of TOCN decreased the water flux due to increased thickness of the bottom layer, porosity and average pore size (Kong *et al.* 2014). This was also similar to the study of Visanko *et al.* (2014) where 2,3-dicarboxylic acid CNF membrane with thicker barrier layer (3.35 μm) produced lower flux compared to the other two thinner barrier layers (0.85 μm and 1.65 μm).

Surface Functionalization. The presence of surface hydroxyl groups enable the possibilities for surface functionalization (oxidation, esterification, etherification, radical grafting, addition, etc.) of CNF and CNC. In the study of Yang *et al.* (2014), a CNF composite was made from thiol-modified CNF embedded in an electrospun PAN nanofibrous scaffold for the adsorption of chromium (VI) and lead (II). Modified CNF with thiol groups induced the formation of Cr(VI)- thiolate and Pb(II) thiolate complex on the surface, thus, producing high adsorption capacities for both Cr (VI) (87.5 mg g⁻¹) and Pb (II) (137.7 mg g⁻¹) (Yang *et al.* 2014). In a separate study, Wang *et al.* (2013) also made a CNF composite membrane with polyvinyl amine (PVAm-CNF). The presence of positively charged groups on the surface of the PVAm-CNF membrane yielded an efficient adsorption not just for negatively charged Cr (VI) ions (100 mg g⁻¹) and Pb (II) ions (260 mg g⁻¹) but also for MS2 viruses (LRV 4) (Wang *et al.* 2013). However, these

charge interactions are dependent on the properties of the pollutant and the pH of the solution. Grafting of sulphate groups onto the surface of CNC by acid hydrolysis imparts a negative charge on the surface. A nanoporous membrane with sulphate-CNC as functional entity and chitosan as matrix successfully removed positively charged dyes (**Figure 3**) such as Victoria Blue 2B (98 %), Methyl Violet 2B (84 %) and Rhodamine 6G (70 %) through hydrogen bonding and electrostatic interaction (Karim *et al.* 2014).

Most of the surface modification of CNF is done before the fabrication of the membrane. A new concept was introduced by Karim *et al.* (2017), where the functionalization of CNF was done in situ after fabrication to enhance the adsorption of metal ions from aqueous medium. Karim *et al.* (2017) fabricated two-layered membranes with different combinations of cellulose sludge (SL), CNF from cellulose sludge (CNFSL) and CNC from bioethanol (CNCBE), unoxidized and in situ TEMPO-oxidized. These layered membranes were as follows: SL/CNCBE, SL-CNFSL/ CNCBE, SL/TEMPO-CNCBE, and SL-CNFSL/TEMPO-CNCBE. The resulting surface modification after the in situ TEMPO-oxidation showed an increase in negative surface functionality of the membranes thereby providing higher adsorption of heavy metals ions such as Ag(I) (0.87 mg g⁻¹), Cu(II) (374 mg g⁻¹) and Fe(II) (456 mg g⁻¹) compared to membranes without functionalization (Karim *et al.* 2017). Even without TEMPO-oxidation, the SL-CNFSL/CNCBE membrane resulted in better adsorption of metal ions compared to membrane without CNF SL as support layer (SL/CNCBE membrane).

Mechanical strength. High mechanical strength indicates higher capability to withstand larger membrane pressure level allowing greater operational flexibility (Spellman 2015). Addition of CNF or CNC to membranes enhances the mechanical property such as increased Young's modulus and slight reduction in strain-at-break that was associated to hydrogen bonding of cellulose (Carpenter *et al.* 2015). Karim *et al.* (2017) fabricated multilayered nanocellulose membranes using vacuum filtration of CNF suspensions followed by dip coating with CNCs having sulfate or carboxyl surface groups that showed high tensile strength of 95 MPa. The work of Ma *et al.* (2014), showed that the Young's modulus and the ultimate tensile strength of CNC-based membranes (375 ± 15 MPa and 14.3 ± 0.4 MPa) were higher than the commercial membrane (GS0.22) and two times higher than the unmodified membrane containing only electrospun nanofibrous scaffolds. The elongation-to-break ratio of commercial membrane (9.4 ± 1.0 %) was lower than those

with nanofibrillar membranes (~23.0 ± 3.0 %). Kong *et al.* (2014) showed in their study that increase of TOCN in CTA also increased the tensile strength and elongation at break. The CTA membrane alone with a tensile strength of 4.64 MPa and 6.0 % elongation at break were greatly increased up to 6.1 MPa and 88 %, respectively, with the addition of 2.5 wt. % TOCN content (Kong *et al.* 2014).

Reusability. Another characteristic of CNF and CNC for membrane applications is their reusability. High regeneration efficiency is an important parameter of membrane performance. Membranes were subjected to desorption tests to determine their regeneration efficiency. EDTA solution (Yang *et al.* 2014), HCl (Wang *et al.* 2013, Yang *et al.* 2014) and/or NaOH (Wang *et al.* 2013) are used to remove lead ions from used CNF-membranes to release metal ions from the membrane surface. After three regeneration cycles, the modified CNF membrane had still the removal efficiency of 93 and 95% of the original removal efficiency for Cr (VI) and Pb (II) (Yang *et al.* 2014), respectively. On the other hand, Wang *et al.* (2013) observed that by using HCl or NaOH solution (depending on the charges), the Pb (II) and Cr (VI) adsorbed on the membrane could still be removed up to 100 % illustrating complete reusability of CNF-membranes using HCl or NaOH solution.

Antibacterial. Another important property of membrane materials is the ability to exhibit bacterial resistance, i.e. their ability to prevent growth of bacteria on the surface and deterioration of the membrane after removal of bacteria. Some previous studies have stated that oxidized cellulose exhibit bacteria inertness due to carboxylate/aldehyde groups on the surface as well as the high degree of crystallinity (Dineen 1976, Ma *et al.* 2014, Hassan *et al.* 2017). TOCN-based thin-film composite membranes fabricated by Ma *et al.* (2014) showed high bacteria inertness with up to 5.5x10⁵ cfu ml⁻¹ of *E. coli* at 37°C for three days maintaining approximately 99.9% the rejection ratio of oil/water. The CNF/AC (activated carbon) membrane from the study of Hassan *et al.* (2017) also showed no growth of bacteria (*E. coli* and *S. aureus*) after incubation for 24 hr at 37°C.

High Crystalline Degree. Chemical resistance is also an important characteristic that needs to be considered for membrane materials as they should be able to withstand harsh chemicals. Cellulose is known to have both amorphous and crystalline regions. The degree of crystallinity of cellulose has a strong influence on its chemical properties due to its ordered chain arrangement that obstruct them to be penetrated by chemicals (Batista *et al.* 2016, Rizkiansyah *et al.* 2016). Ma *et al.* (2014) showed

in their work that CNF based thin-film composite membranes were inert to oily waste water composed of oil mixture, hydrazine, chlorobenzene, acetone and phenol while maintaining the rejection ratio above 99.4%. This is due to the crystalline and thermally cross-linked structure of CNF to the matrix. Although CNC is highly crystalline, there is still limited data documenting the chemical resistance of CNC based membranes.

Applications of CNF and CNC Based Membranes in Water Purification

These literatures which includes a summary of list of nanocellulose membrane demonstrated in the recent literature for water purification, shows that nanocellulose are used as an important functional additive to improve the performance, pore structure, mechanical stability and adsorption capacity of the membrane due the characteristics of nanocellulose mentioned in previous section (**Table 4**).

This section focused on some applications of CNF and CNC membranes especially on treating actual samples as it will be helpful for developing new

nanoporous materials for quick, economic, and high-performance water purification.

Removal of Organic Contaminants. Organic pollutants such as pesticides, hydrocarbons, phenols, oils, and pharmaceuticals are known to reduce the amount of dissolved oxygen in water in their oxidative decomposition process, thus endangering the aquatic organisms. Previous works of *Karim et al. (2014)*, *Goetz et al. (2018)*, *Liu et al. (2019)* and *Soyekwo et al. (2016)* showed success in removing about 70% to 90% dyes such Victoria Blue 2B, Direct Red 16, Methyl Violet 2B, and Rhodamine 6G from water by preparing a nanoporous membranes with CNC as functional entity in chitosan, CNC-impregnated electrospun cellulose acetate and CNF membrane, respectively. These membranes adsorbed dyes mainly through electrostatic attraction and hydrogen bonds (*Karim et al. 2014*, *Soyekwo et al. 2016*, *Goetz et al. 2018*, *Liu et al. 2019*).

Natural organic matter were identified as one of the main components responsible for membrane fouling as it consists of various fractions, among which humic-like substances, and biopolymers (*Chen et al. 2015*; *Shao et al.*

Table 4. Process and characteristics of Cellulose Nanofibrils and Cellulose Nanocrystals based membranes for water purification.

Raw material of nanocellulose	Membrane production process	Membrane	Membrane thickness (μm)	Pore size	Target pollutant	Flux	Adsorption (mg g^{-1})	Percent rejection	Reference
Cellulose nanocrystal powder	Freeze-drying	CNC-Ag	0.149-0.236		Na_2SO_4	$25.4 \text{ L h}^{-1} \text{ m}^{-2} \text{ bar}^{-1}$		99.1%	(<i>Xu et al. 2020</i>)
Commercial CNF	Vacuum filtration and suspension	GO-CNF	19.3	0.107-0.298 μm	Dyes	$18123 \text{ L h}^{-1} \text{ m}^{-2} \text{ bar}^{-1}$		Victoria blue – 98.8 % Methyl violet – 97.6 % Rhodamine 6G-92.3%	(<i>Liu et al. 2019</i>)
Filter paper	Non-solvent induced phase separation (NIPS) method	a. CNC/PVDF b. DDBAC/CNC/PVDF, c. ZnO/CNC/ PVDF d. GO/CNC/PVDF	a. 0.018 b. 0.023 c. 0.025 d. 0.028		Humic Acid	a. $75 \text{ L h}^{-1} \text{ m}^{-2} \text{ bar}^{-1}$ b. $130 \text{ L h}^{-1} \text{ m}^{-2} \text{ bar}^{-1}$ c. $175 \text{ L h}^{-1} \text{ m}^{-2} \text{ bar}^{-1}$ d. $230 \text{ L h}^{-1} \text{ m}^{-2} \text{ bar}^{-1}$		a. 98% b. 95% c. 95% d. 92%	(<i>Zhang et al. 2019</i>)
Cellulose Nanocrystals	Phase inversion	PES-amine functionalized CNC	150		Copper ion Direct red 16	$25 \text{ kg h}^{-1} \text{ m}^{-2}$	-	Copper ion – 90% Direct red 16– 99%	(<i>Rafeian et al. 2019</i>)
CNC from Unbarked wood	Impregnation into electrospun	CA-CNC	52–116		0.05–2.0 μm particles, Victoria blue	$500\text{--}5000 \text{ L h}^{-1} \text{ m}^{-2} \text{ bar}^{-1}$		0.05–2.0 μm particles – 56%; Victoria blue 2B -99%	(<i>Goetz et al. 2018</i>)
Commercial Cellulose Microcrystalline cellulose	Spray coating Interfacial polymerization	PAN-TEMPO-CNF PA/CNC/PES	0.2-0.47	0.06-0.15 μm	Dextrans Na_2SO_4	$34\text{--}230 \text{ L h}^{-1} \text{ m}^{-2}$ $60 \text{ L h}^{-1} \text{ m}^{-2}$ $204 \text{ L h}^{-1} \text{ m}^{-2}$		62-75% 96.7 %	(<i>Wang et al. 2017</i>) (<i>Wang et al. 2017</i>)
Cellulose sludge and unbarked wood form bioethanol plant	Vacuum filtration, deep coated with CNC _{BE} , acetone treated and TEMPO-oxidized	SL-CNF _{SL} /TEMPO-CNC _{BE}	201-206	0.56–0.60 μm	Ag^+ Cu^{2+} $\text{Fe}^{3+}/\text{Fe}^{2+}$	$55 \text{ L h}^{-1} \text{ m}^{-2}$ MPa^{-1} $77 \text{ L h}^{-1} \text{ m}^{-2}$ MPa^{-1}	Ag^+ - 0.87 Cu^{2+} - 374 $\text{Fe}^{3+}/ \text{Fe}^{2+}$ - 456		(<i>Karim et al. 2017</i>)

Table 4. Process and characteristics of Cellulose Nanofibrils and Cellulose Nanocrystals based membranes for water purification. (cont.)

Raw material of nanocellulose	Membrane production process	Membrane	Membrane thickness (μm)	Pore size	Target pollutant	Flux	Adsorption (mg g^{-1})	Percent rejection	Reference
Palm fruit stalks pulp	Drying – alcohol exchanged	Upper layer: CNF+PAE, Lower layer: AC+TOCN+PAE	0.496–0.564	0.061-0.172 μm	<i>E-coli</i>	158 L h ⁻¹ m ⁻²	-	99 %	(Hassan et al. 2017)
Commercial α -cellulose powder	Vacuum filtration	CNF			Ferritin Gold Methylene blue	0.952–2.2 L h ⁻¹ m ⁻²		Ferritin-84-93% Gold-90-94% Methylene blue-99%	(Soyekwo et al. 2016)
Cellulose nanofiber from eucalyptus kraft pulp	Phase inversion	CA-CNF		-	Whey, permeated strawberry (SJ) and raspberry (RJ) juices total solids (TS) and Soluble solid (SS)	940 L h ⁻¹ m ⁻²	-	SJ (TS) – 41 % (SS) – 34 % RJ (TS) – 39 % Whey (TS) – 3 % (SS) – 4 %	(Battirola et al. 2017)
Cellulose Sludge and unbarked wood from bioethanol plant	Vacuum filtration, deep coated with CNC and acetone treated (hot pressing)	CNC _{BE} -CNF	176	50-100 Å (max)	Ag ⁺ , Cu ²⁺ Fe ³⁺ /Fe ²⁺	15-25 L h ⁻¹ m ⁻² MPa ⁻¹	Ag ⁺ -0.023 Cu ²⁺ -33 Fe ³⁺ / Fe ²⁺ - 55	94 % 99 % 100 %	(Karim et al. 2016)
Commercial MCC (microcrystal cellulose)	Phase inversion	CNC-PES		-	NaCl MgSO ₄ Acid Orange Whey protein	~33 kg h ⁻¹ m ⁻²	-	NaCl -16.1 % MgSO ₄ - 42.1 % Acid orange- 79.1 % Whey protein-99 %	(Daraei et al. 2016)
Dry wood pulp cellulose	Impregnation into electrospun PAN scaffold	Thiol functionalized CNF-PAN	200	0.43 μm (mean)	Cr (VI) and Pb (II)	1000 L h ⁻¹ m ⁻²	Cr(VI) - 87.5 Pb(II) - 137.7	-	(Yang et al. 2014)
Bleached birch chemical wood pulp	Vacuum Filtration and solvent exchange	DCC nanofibril-commercial membrane	0.85-3.35	0.017-0.051 μm .	Dextrans	1710 L h ⁻¹ m ⁻²	-	74-80 %	(Visanko et al. 2014)
Cellulose (Biofloc 92 MV, wet, 22 wt% of wood pulp)	Impregnation into electrospun PAN scaffold	CN-TFNC		0.024 μm (mean) 0.0546 μm (max)	0.10 μm microsphere, Oil/water, Sodium alginate solution	199-303 L h ⁻¹ m ⁻²	-	0.10 μm - 97 % Oil/water – 99.8 % Sodium alginate solution - 96.3 %	(Ma et al. 2014)
Softwood bleached kraft pulp	Phase inversion	CTA-TOCN		0.047-0.051 μm	Protein	224.68 L h ⁻¹ m ⁻²	-	75 %-95 %	(Kong et al. 2014)
Non-dried cellulose residue	Freeze-drying	CNC-chitosan	250-270	0.013 μm	Dyes	64 L h ⁻¹ m ⁻² MPa ⁻²	-	Victoria blue – 98 % Methyl violet – 90 % Rhodamine 6G-70%	(Karim et al. 2014)
Wood pulp	Impregnation into electrospun PAN scaffold	PVAm grafted CNF	40	0.38 μm (mean) 0.73 μm (max)	Bacteria, Virus, Cr (VI) and Pb (II)	~1300 L h ⁻¹ m ⁻² psi ⁻¹	Cr (VI) - 100 Pb (II) - 260	Bacteria-100 % Virus-LRV4	(Wang et al. 2013)
Micro crystalline cellulose	Impregnation into electrospun PAN scaffold	CNC-PAN	45	0.22 μm (mean), 0.41 μm (max)	Positively charged crystal violet dye <i>E-coli</i> , <i>B. diminuta</i> and MS2	59 L h ⁻¹ m ⁻² kPa ⁻¹	Positively charged crystal violet dye - 68	<i>E-coli</i> – LRV 6 <i>B. diminuta</i> – LRV 6 MS2 - 2	(Ma et al. 2012)
Cellulose pulp	Phase inversion	CNC-PVDF		0.039-0.049 μm	Bovine serum albumin solution	230.8 L h ⁻¹ m ⁻²	-	92.5 %	(Bai et al. 2012)
Wood pulp	Impregnation into electrospun PAN scaffold	CNC-PAN	6.5	0.0546 μm	MS2 bacteriophage Oil/water	55.6 L h ⁻¹ m ⁻² psi ⁻¹	-	MS2 LRV - 1.6-3.7 Oil/Water – 99.5 %	(Ma et al. 2011)

2019). As previously discussed, CNF and CNC showed great performance in terms of reducing the membrane fouling. Zhang *et al.* (2019) investigated the anti-biofouling performance of DDBAC/CNC, ZnO/CNC and GO/CNC nanocomposites modified polyvinylidene fluoride (PVDF) ultrafiltration membranes for practical application in micro-polluted source water purification. Among these hybrid membranes, GO/CNC/PVDF exhibited an enhanced perm-selectivity with a water flux of 230 L/(m²-hr bar) and humic acid rejection of 92%. There is also an improvement of on its antibacterial activity (93%) and antifouling performance (flux recovery rate (FRR) of >90%), due to the optimized pore structure, higher surface roughness, incremental hydrophilicity and electronegativity (Zhang *et al.* 2019).

In another study, Cheng *et al.* (2019) investigated the removal and antifouling performance of CNT and CNF coated membranes with typical NOM (humic acid (HA), bovine serum albumin (BSA), and sodium alginate (SA)) and its application in natural surface water collected from Yingxue lake in Jinan, China. Generally, in actual waters, humic substances, proteinaceous and polysaccharides were coexistence, and the surrogate foulants may not be relevant to actual water bodies. The rejection rate of HA and BSA by virgin membrane were 46.4%, and 15%, and were increased to 63-69% and 20-27% for CNFs coated membranes, respectively. By contrast, CNTs coated membrane showed an increase rate of 82-90% for HA and 60-90% for BSA. For SA removal, both CNTs and CNFs coated membranes showed very limited improvement compared to the virgin membrane. During its application in the natural surface water treatment, DOC concentration were reduced from 6.24 to 4.88 mg L⁻¹ after filtration by virgin UF membrane, and further decreased to 2.83 and 4.2 mg L⁻¹ during the presence of pre-deposited CNTs and CNFs layers, respectively. During HA fouling, the minimum coating mass of 6 g m⁻² exhibited the best mitigating performance for CNTs and CNFs coated membrane, whereas for BSA fouling, the best performance was obtained at the maximum coating amount of 50 g m⁻². Furthermore, the transition of fouling mechanisms during filtration of natural surface water were significantly delayed from pore blocking to cake filtration when using pre-deposited coating layers (Cheng *et al.* 2019). However, CNTs coating layers exhibited better performance compared to CNFs coated membranes in terms of removal efficiency and alleviation in reversible and irreversible fouling. CNFs exhibited limited effect on irreversible fouling control.

Removal of Inorganic Contaminants. As mentioned in the previous section, the abundant functional groups

on nanocellulose can provide excellent adsorption sites which can remove inorganic contaminants as well as the organic contaminants.

The adsorption contact time directly affects the membrane lifetime and the adsorption time efficiency. The adsorption isotherm can be used to illustrate the interactions between adsorbents and adsorbates, as well as the adsorption capacity of adsorbents (Yang *et al.* 2014). The most common isotherm for sorption of cellulose is the Langmuir isotherm, which can be expressed by Equation (1):

$$q_{eq} = \frac{Q_{max}bC_e}{1+bC_e} \quad (1)$$

where C_e (mg L⁻¹) is the equilibrium concentration of the pollutant in the solution; q_{eq} (mg g⁻¹) is the (equilibrium) adsorbed amount at this solution concentration; Q_{max} (mg g⁻¹) is the maximum adsorption capacity per gram of sorbent; and b (L mg⁻¹) is the Langmuir constant related to the adsorption energy of the system (Voisin *et al.* 2017). Adsorption capacity of membrane for ions can also be calculated using Equation (2):

$$q_{eq} = \frac{(C_0 - C_e)V}{W} \quad (2)$$

where C_0 and C_e are the initial and equilibrium solution concentrations (mg L⁻¹), respectively, V the volume of the solution (L) and W the membrane weight (mg) (Karim *et al.* 2016). In order to examine the controlling mechanism of the biosorption process such as mass transfer and chemical reaction, kinetic models are used to test the experimental data (Bayramoglu *et al.* 2002). A plot of t/q_t where q_t (mg g⁻¹) is the amount of metal absorbed at time vs. time (t) should give a linear relationship for the applicability of the second-order kinetic where the chemisorption process is the rate determining step (Bayramoglu *et al.* 2002, Voisin *et al.* 2017). In a true first order process $\log q_{eq}$ should be equal to the intercept of a plot of $\log (q_{eq} - q_t)$ against t . However, kinetic studies are currently insufficiently presented in literature (Voisin *et al.* 2017). Improvement of removal and adsorption capacities has been noticed after the addition of CNF or CNC on membrane materials. Although in some cases, the water permeation flux decreases after addition of CNF or CNC, the effect on the adsorption capacities and removal of pollutants were not affected (Karim *et al.* 2014, Wang *et al.* 2017).

Recent studies showed that the use of CNF membranes, having highly porous and highly interconnected pore structures allows the water molecules to permeate

easily. However, the highly porous structures of CNF membranes produced from the electrospinning process cause the metal ions to easily escape from the membrane layer despite having superior water permeability. To overcome this problem, *Chitpong and Husson (2017)* developed a high-performance ion-exchange membranes for cadmium recovery from impaired waters by using subsequent modified cellulose acetate nanofiber membrane. Poly (acrylic acid) was grafted to the surfaces of individual nanofibers that had been modified with poly(glycidyl methacrylate). The separation results showed a superior Cd^{2+} adsorption capacity of up to 160 mg g^{-1} which are comparable to traditional ion-exchange media. The specific area of electrospun nanofiber membranes was measured to be 1.69 times higher than the PAA commercial RC membranes (Whatman RC 60). The high binding capacities were also achieved due to the solvents used for PAA grafting (*Chitpong and Husson 2017*). In another study, cellulose nanofiber mat was grafted with PAA and poly(itaconic acid) (PIA) for selective removal of heavy metals from impaired waters. Very high permeability was obtained and the adsorptive capacity of the membranes was 162 mg g^{-1} and 222 mg g^{-1} for Cd(II) and Ni(II), respectively. Furthermore, the CNF with PAA and PIA membranes were very selective for Cd over Ni from the impaired water. The higher metal-polymer complex stability of Cd-PIA over Cd-PAA was found to be a factor for achieving high Cd binding capacities (*Chitpong and Husson 2017*). Meanwhile, *Stenina et al. (2020)* showed that the type of grafting polymer, the solvents to be used in grafting, and inclusion of CNF membranes are factors affecting the binding capacities of membrane.

In the study of *Yang et al. (2014)*, a maximum adsorption of 80 mg g^{-1} for Cr (VI) and 125 mg g^{-1} Pb (II) within 15 and 20 min, respectively, was obtained due to fast formation of Cr-thiolate complex and Pb-thiolate complex through the chelating reaction of SH and Pb(II) using thiol-modified CNF membrane. In another study, *Venäläinen and Hartikainen (2017)* did a meaningful work on preparation of anionic nanofibrillated cellulose to adsorb quickly, efficiently, and simultaneously both cations (Fe, Al, Mn, Ni, Mg, and Na) and anions (SO_4^{2-}) from authentic acidic mining water. Anionic CNF gels with different concentration, after allowing to react for 10 min with mining water, efficiently co-adsorbed both cations and anions. The retention of cations by ionic CNF was suggested to result from electrostatic adsorption of positively charged ions by the deprotonated adsorbent surface through the formation of metal-ligand complex. The subsequent formation of net positive charge on the adsorbent surface enabled the electrostatic adsorption

of anions (SO_4^{2-}) (*Venäläinen and Hartikainen 2017*). However, for nanocellulose membranes, its application on mining water are still limited and there are a lot of challenges and problems still need to address (*Agboola 2019*).

CONCLUSION AND RECOMMENDATIONS

Both CNF and CNC, with the abovementioned properties, provide new perspectives in generating membranes that can be used for water purification. The addition of CNF and CNC enhances the properties of the membranes. The enhancement was associated to the membrane's hydrophilic characteristics, charges and pore size distribution. Functionalized CNF and CNC membranes have been shown to remove heavy metal pollutants, viruses, dye and bacteria from aqueous solution. Introducing charge onto the surface of cellulose increases the surface area to volume ratio.

Membrane fabrication using different processing techniques were also briefly discussed in this review. However, addition of CNF or CNC shall be controlled; and pore structure, accessibility to the functional entities for interactions with contaminants without compromising membrane flux, and rejection capacity are factors that shall be considered. Heavy loading of CNF and CNC could decrease the water flux of the membrane. Hence, it is important to optimize the amount of CNF or CNC. Kinetic models are also useful to determine the capacity of the biosorbent (CNF or CNC) to adsorb and release contaminants, but this information is currently insufficiently presented in literature.

So far, most investigations on nanocellulose membrane production are only on a laboratory scale. Though, efforts been made on investigating the performance of nanocellulose membrane by using actual water samples. The CNF and CNC show antibacterial activity due to the excellent hydrophilicity, cytotoxicity, and electronegativity, which makes them a great additive on an anti-biofouling membrane. However, further studies are still needed especially on reducing irreversible fouling. Also, the solvents used and the type of grafting the polymer played an important role in binding capacities.

Despite of all aforementioned advantages of using CNF and CNC for membranes, there are challenges that could hinder their applicability and performance. These include cost efficient methods for large scale production, mechanical properties, lifetime, disposal routes and the degradation after interaction with bacteria for some time. Materials with CNF are one group of recyclable

materials that exhibit impressive biodegradable properties. However, there are still some concerns on its degradability after interactions with bacteria. Studies have shown that CNF and CNC as additives in membranes might not degrade if they are stably contained in the bulk of polymers. Hydrolysis is the initial rate-limiting step of cellulose biodegradation. Metagenomics could also be a key to provide insights into fundamental aspects of cellulose biodegradation. Biodegradable studies indicated that the complete degradation of membranes coated with CNC is within 15 to 30 days in soil under temperature variation (Karim *et al.* 2017). There is a need to further investigate the selectivity of the membranes in the presence of more complex water streams; therefore, introduction of more functionalities is essential. Given the wide range of benefits that CNF and CNC offer, comprehensive researches are needed to further the development of the high strength, thin, high flux and with high selectivity membrane leading to the commercialization of this promising material as membrane for water purification.

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