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REVIEW

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Nanocellulose as a sustainable material for water purification

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Abstract
The demand for purified water has been increasing day by day. More feasible technologies, including membrane filtration, adsorbents, and so forth have emerged out to be more efficient and cheaper over conventional industrial methods. Nanocellulose, being biodegradable, nontoxic, and sustainable nanofiller exhibits excellent mechanical properties, high aspect ratio, high surface area, and more importantly tunable surface chemistry; is a potential candidate to be employed for water purification. Composite membranes and films for water filtration, constituting of biopolymers have gathered immense interest lately. Compared with its unmodified form, the functionalized NC enhances the compatibility with the matrix and readily forming strong network structures; essential for the formation of channels for better adsorption of impurities and higher water flux. This review highlights some of the recent studies dedicated to making and testing of nanofiltration membranes prepared using nanocellulose and its different functionalized derivatives.

KEYWORDS
membrane, nanocellulose, nanocomposites, purification, water

1 | INTRODUCTION

The declining quality of water resources is a major problem faced by humanity in the 21st century, due to increasing industrialization and urbanization releasing organic and inorganic pollutants into water. Pollutants like heavy metal ions, dyes, oils, salts, and so forth are hazardous to both human health and the environment. The rapid growth of the population has led to an increase in the demand for freshwater, especially for food production, as agricultural irrigation alone requires water which amounts to about 70% of the freshwater reserves around the globe.[1] Hence, the decontamination of water has found itself to be an urgent issue. Various water remediation techniques such as membrane separation, ion exchange, photocatalysis, adsorption, and electrochemical treatment have been studied.[2,3] Some of these methods, however, require substantial capital, and their use is restricted due to certain issues. These issues may vary with different water treatment methods in different industries. Considering the food industry, removal of microorganisms, low water flux, and high energy consumption associated with the reverse osmosis and nanofiltration (NF) process, are certain issues that one must be concerned of ref. [4, 5] Studies in the recent past have focused on preparing bioflocculants based on modified polysaccharides.[6] Flocculants are commonly used materials for the removal of contaminants from wastewater by accelerating agglomeration of these contaminants, causing sedimentation, followed by contaminant removal.[7] Through the years, polysaccharides have become an attractive alternative option for fabricating a wide variety of high performance and low cost adsorbent membranes, attributed to the ease of their chemical modification.[7] Membrane-based water treatment eliminates the requirement of chemical additives, no phase change is involved during the process, and the concept is simple and easy to implement. Thus, membrane water treatment plays an important role in areas such as seawater and brackish water desalination, and wastewater treatment and reuse.[8,9] Membranes and membrane filtration processes, have become an essential part of the food processing layout, where about 20–30% of these membranes have found use in the food industry.[10–12] Because of its controllable pore size, convenient operation, and low energy consumption, membrane filtration is a promising separation technique.[13–19]

Of late, nanomaterials have found unique applications in the preparation of nanocomposites and developed new fields of interest in scientific studies. For instance, one of the most recent studies by Tanwar et al.,[20] highlights the use of nanoscale nickel zinc multiferrites for photocatalytic decontamination of water. Nanomaterials/nanofillers provide enhanced physicochemical resistance, adsorption of contaminants, and so forth for water purification.[21–23] Bio nanofillers having good compatibility, accessibility, affordability, are easily producible, and are nontoxic. These are some of the reasons why studies have inclined toward the synthesis of nano-biomaterials.[24–26] Cellulose is one such material which fulfills all the mentioned criteria to be a perfect subject for research. Cellulose, however, lacks in the ability of water decontamination due to low chemical and physical stability, and hence, low adsorption[27]. Whereas, a vast number of hydroxyl groups present on the surface of nanocellulose (NC), a derivative of cellulose, allows its surface modification using a variety of chemicals, and tuning the properties of the composites as per requirement.[28–30] NC is obtained through a series of processes including mechanical fibrillation of cellulose, followed by chemical treatment for purification, or removal of lignin, hemicellulose, and so forth. Cellulose nanocrystals (CNCs) are obtained by hydrolysis of alkali-treated, delignified cellulose fibers, using strong acids. Whereas, cellulose undergoes chemical pre-treatment followed by shearing processes, to yield nanoscale cellulose fibers, commonly known as cellulose nanofibrils (CNFs). Unlike, CNCs and CNFs, bacterial nanocellulose (BNC) is obtained by bacterial synthesis, thus being free from lignin or hemicellulose. Hence, BNC is regarded as the purest NC available. In the recent past, studies have

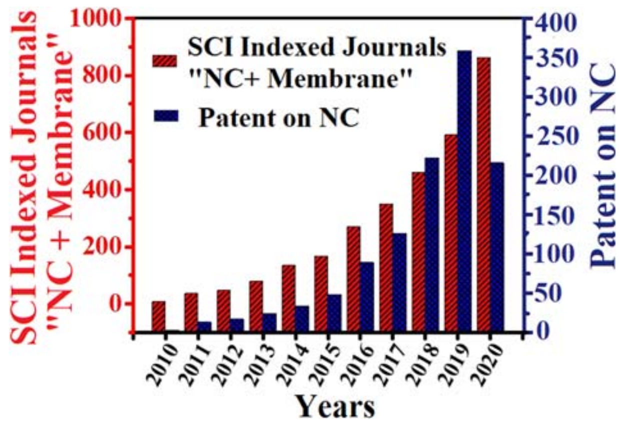

FIGURE 1 Growing importance of NC reflects in the increasing number of SCI-indexed publications and patents granted over the last decade

been dedicated to preparing nanocomposite membranes based on the abovementioned materials. The so-formed composite membranes have been employed in tests, to get rid of various contaminants in water such as arsenic, vanadium, mercury, liquid dyes, phenol, chromium, lead, natural organic matter, and so forth.[31–34] Figure 1 shows the number of SCI-indexed journals published employing NC in water filtration/purification applications. The number of patents on NC granted demonstrates its overall growing importance in coming future (SciFinder Scholar database, August 2020).

This review highlights some of the recent studies in the production of NF membranes based on chemically modified NC. These studies mainly the advantages associated with the interaction of NC with the different organic matrices, in terms of better adsorption of unwanted contaminants, including heavy metal ions, toxic dyes, salts, and oils, increased efficiency of purification, higher water flux, and so forth.

2 | NANOCELLULOSE BASED MEMBRANES

A new method for pollutant removal is membrane separation with no requirement for phase change, reduced energy consumption, ease of particle separation even from extremely dilute solutions, and higher efficiency.[35] Color separation in membrane filtration processes is usually carried out with NF and reverse osmosis, among which NF membranes seem to provide the best performance in separation, resulting from their nanoporous structure through which, most organic matter cannot pass.[36,37] Due to advantages such as a high degree of flexibility, relatively low cost, smaller footprint, and straightforward pore-forming mechanism, polymeric membranes are currently the most widely used membrane type for water purification. Nanocomposite membranes consisting of dispersed nanomaterials in a polymer matrix are used in liquid–solid, gas–gas, and liquid–liquid separations. For this, NCs show great potential, owing to their biodegradability, high specific strength, stiffness and aspect ratio, renewability, low price as well as good thermal stability. Sequential vacuum filtration is a prominent method employed for the manufacture of nanocellulose membranes. It is a standard technique used to separate a solid–liquid mixture for the purpose of retaining the solid. It is similar to gravity filtration where the mixture is separated by using a filter, the differentiating factor being that vacuum filtration is aided by a vacuum pump beneath the filter paper separating the mixture. Vacuum filtration is faster than gravity filtration, because the solvent or solution and air is forced through the filter paper by the application of reduced pressure. It is also more efficient at removing residual liquid, leading to a purer solid. However, due to the force of suction fine crystals may be pulled through the filter paper pores, leading to a quantity of material that cannot be recovered from the filter paper, and possibly an additional quantity that is lost in the filtrate. When it comes to nanocellulose in particular, nanocellulose membranes (along with a composite material) are usually prepared along vacuum filtration of an aqueous nanocellulose dispersion on a filter paper substrate. Drying methods further influences the filtration performance of the membranes. The membranes seem to exhibit better rejection rates with lower water flux, with a greater NC aspect ratio and an increasing amount of added NC.

Along with membranes, domestic water filtration systems, being very effective are used throughout the world. This system involves the placement of a filter at the tap outlet while using the pressure of the water that comes out of the pipe.[38–40]

Apart from CNFs and CNCs, their altered derivatives have also been studied abundantly for adsorption of impurities such as heavy metals, dyes, and organic compounds from the water via electrostatic interactions.[41–44] CNF was found to be an ideal material for membranes, for removing charged pollutants from the aqueous environment through electrostatic interactions along with size exclusion due to its hydrophilicity, tailorable surface geometry, and network formation properties. However, it has drawbacks such as feeble wet stability of CNF based networks, low water permeation,[45] and lack of functionality, unless the surface is modified.[46] These membranes have been studied for the removal of various contaminants including heavy metal ions, toxic dyes, salts, oils, and so forth Table 1. Mentions the results in terms of efficiencies of such membranes in the removal of contaminants.

2.1 | Removal of heavy metal ions

Heavy metal pollution poses a serious threat to human health. For instance, the use of copper in various industries is associated with the contamination of water sources, including groundwater sources,[51–53] possibly resulting from its leakage. Excessive intake of copper ions in the human body would have serious implications.[54] The same is applicable for metals such as silver, iron, chromium, and other commonly used metals in industries. Many processes have been adopted in the past to clear heavy metal water pollution such as chemical precipitation, photocatalysis, ion exchange, physical adsorption,[55–60] and biological treatment, of which physical adsorption is heavily favored due to its easy process and environment friendliness.[61,62] Jalali et al.[63] and Johari et al[64] used cellulose derived from sunflower stalks and coconut husk wastes, for the adsorption of cadmium and lead metal ions, and mercury metal ions, respectively. Both the groups obtained satisfactory results in terms of metal ion adsorption. On the other hand, Bediako et al.[65] utilized waste lyocell fabric to prepare carboxymethyl cellulose adsorbent via crosslinking reactions and carboxymethylation, to remove Cadmium ion (Cd^{+2}). This produced adsorbent displayed an ~17 times higher metal uptake. It was realized that the nanofiber suspension could remove a significant amount of Cd^{2+} ions, even at low nanofiber concentration, in under 5 min. A cellulosic adsorbent was prepared by Sun et al[66] by halogenating microcrystalline cellulose followed by its functionalization with pyridone diacid for removing Pb^{+2} and Co^{+2}. Apart from these, several studies revolve around the use of different chemical materials to modify CNCs and utilize them as adsorbents for water purification.[67–72] An investigation was conducted by Rafieian et al,[42] involving the preparation and characterization of a membrane, consisting of (3 aminopropyl)triethoxysilane or APTES-modified CNC (MCNC) and a nonwoven substrate at the bottom for removing heavy metals like copper ions present in water. It was observed that MCNC was responsible for the improvement of fouling resistance and water flux of the polyethersulfone (PES)/MCNC membrane and increased efficiency in metal ion removal. Figure 2A gives a graphical representation of how the copper removal percentage varies with time for various MCNC percentages (for reference—PES-0.1 denotes 0.1 wt% MCNC added to PES, and so on.) within the PES/MCNC membrane. Clearly, the membrane containing 1% MCNC, had the highest percentage of Cu^{+2} ions removed, reason being the high dispersion of MCNC). This proved that the presence of MCNCs in the

TABLE 1 Tabulation of different researches in membrane filtration and their efficiencies in terms of contaminant removal

Serial no.	Composite	Contaminants	Removal efficiency	Reference
1.	PES/1%MCNC	Copper ions	90%	[42]
2.	PCL50:CNF50	Chromium	99%	[43]
		Iron	75%	
3.	TEMPO oxidized-carboxylated NC	Methylene blue	118 mg g^{-1}	[44]
4.	Amino-functionalized CNCs	Acid red GR	555.6 mg g^{-1}	[45]
5.	PDA/BNC	Rhodamine 6 G		[46]
		Methylene blue		
		Methyl orange		
		Lead	16.8 mg g^{-1}	
		Cadmium		
6.	P-MPC/BNC	Methylene blue	4.44 mg g^{-1}	[47]
		Methyl orange	4.56 mg g^{-1}	
7.	Double layered GO/CNF membranes	Victoria blue 2B	98.8%	[48]
		Methyl violet 2B	97.6%	
		Rhodamine 6 G	92.3%	
8.	PES/1%MCNC	Colored dissolved compounds form licorice processing industry	94.2%	[49]
9.	CNC based nanofiltration membrane	Na_2SO_4	98%	[50]
		Mg_2SO_4	96%	

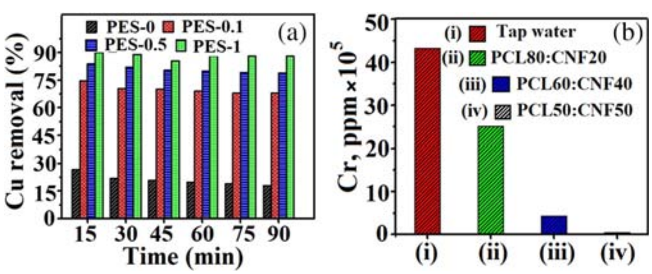

FIGURE 2 (A) Percentage removal of Cu^{+2} ions as a function of MCNC concentration and time.[42] (B) Concentration of chromium metals, passed through membranes having PCL and CNFs in different proportion[43]

membrane structure improved the efficiency of the nanocomposite in copper (II) removal. The PES-1 wt% membrane was further subjected to filtration-regeneration tests, showing a minimum decrease in efficiency of the membrane.

In the recent past, CNFs,[73] which can also be obtained from the industrial crop,[74–77] had been introduced as adsorbing material for different ions in still media.[78–85] Agave bagasse was found to be an abundant source of this lignocellulosic biomass.[86–88] Agave has recently been explored for the production of cellulosic nanocrystals and nanofibers.[43,89–91] Hinestroza et al,[43] prepared electrospun, organic membranes from CNFs obtained from agave bagasse fibers, and polycaprolactone (PCL), mixing both at different *v/v* ratios (denoted by PCL80:CNF20, PCL60:CNF40, PCL50:CNF50). NC has also been reported to be useful in the adsorption of heavy metals due to its negative surface charge. So, these membranes were also expected to be used for selective heavy metal contaminants and to reduce the turbidity and conductivity of the water for human consumption. The electrospun PCL50:CNF50 (vol/vol ratio) membranes not only removed 100% of the conductivity and turbidity but also successfully retained a good percentage of the heavy metals (Cr = 99% and Fe = 75%) that were present in the tap water. Figure 2B shows the decrease in the concentration of Cr ions in tap water, filtered using different membranes. The results of the study displayed that CNF is useful in the production of PCL/CNF electrospun membranes which could possibly find application in eco-friendly filtration systems for water purification.

From CNF, cellulose nano-papers can be prepared which have also found application in membrane processes.[89–91] However, the abovementioned nano-papers seemed too dense, thereby, hindering high water permeance and reducing the efficiency. It, however, seemed possible that the decoration of conventional natural fiber substrate with charged cellulose, would solve the issue and allow high permeance, simultaneously providing adequate contact area for the adsorption of dissolved ions during the passage of water through it. Mautner et al.[92] presented an approach that used natural adsorbent material to penetrate a natural fiber substrate obtained from agave and flax. They prepared a nonwoven substrate from a mixture composed of agave and flax fibers, and incorporated anionic TEMPO [(2,2,6,6-Tetramethylpiperidin-1-yl)oxyl]-oxidized CNFs into it. This provided a high net negative surface charge, supporting the adsorption of cationic metal ions. Over 1200 mg m^{-2} could be adsorbed onto the filters, which corresponded to over 60 mg g^{-1} of adsorption capacity for TEMPO-oxidized-CNF, which was the active adsorptive agent. These properties allowed for the highly efficient application of these membranes in adsorbing heavy metal from aqueous solutions. The permeance of flax agave (FA) filters in comparison with cotton viscose (CV) filters with varying added CNF can be seen in Figure 3.

2.2 | Removal of toxic dyes

Dye pollutants that are organic in nature display anionic, cationic, or nonionic properties. Cationic dyes are adsorbed by employing anionic moieties functionalized NC. Batmaz et al[44] found that carboxylated NC synthesized by TEMPO-mediated oxidation produced an expressively higher dye uptake of cationic methylene blue at pH = 9. As for anionic dyes, cationic NC prepared by successive oxidation using sodium periodate, ensued by a reaction with ethylenediamine showed the maximum adsorption of acid red GR, as studied by Jin et al.[45]

BNC and its composites have been used to manufacture water remediation systems for adsorbing heavy metals, oils, and dyes.[93–98] Gholami et al.[46] utilized

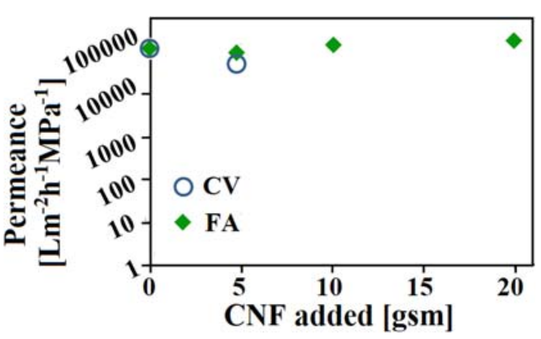

FIGURE 3 Graph showing the permeance values of both cotton viscose (CV) and flax-agave (FA) substrates as a function of the amount of CNF added[87]

BNC and polydopamine (PDA) for fabricating scalable and reusable films (Figure 4A), capable of adsorbing toxic dyes such as rhodamine 6 G, methyl orange, methylene blue, and heavy metal ions (Figure 4). The partnership between a zwitterionic polymer and BNC had not been previously explored for the development of nanocomposites for the removal of organic dyes, before Vilela et al [47] fabricated nanocomposite membranes consisting of BNC and cross-linked poly (2-methacryloyloxyethyl phosphorylcholine) (PMPC) to develop a three-dimensional network. The 2-methacryloyloxyethyl phosphorylcholine (MPC) was the chosen as the nontoxic, polymerizable polymer due to its methacrylic functional group and its zwitterionic phosphorylcholine moiety, which consisted of a trimethylammonium cation and a phosphate anion.[99] This MPC polymer also displayed bioinert, antimicrobial, and antifouling properties,[100–102] as well as a unique hydration state which would be major assets in the reduction of microbial growth in contaminated water. They studied the thermal stability, antibacterial properties, and mechanical properties of various bacteria, as well as the water uptake ability and the removal of organic dyes. They were found to show a dye adsorption amount of 4.44 ± 0.32 mg g^{-1} of methylene blue and 4.56 ± 0.43 mg g^{-1} of methyl orange for the higher zwitterionic polymer (i.e., 79 wt% of PMPC) containing membrane. The results showed that the developed nanocomposite membranes were proficient in the adsorption of cationic and anionic organic dyes. The membranes also displayed good thermal stability up to 250°C, inhibited the growth of certain bacteria, and good mechanical properties.

Several carbon-based materials such as multidoped carbon fibers, graphene oxide, and so forth have been used studies related to the decontamination of wastewater.[103–105] Among these, graphene oxide (GO) sheets have found use as a selective membrane allowing permeation of molecules and ions into the aqueous solutions.[103,106] GO, when prepared from graphite by chemical exfoliation, has recently been discovered to be a nanomaterial full of potential for adsorption owing to its large surface area, extremely reactive surface due to the profuse carboxyl and hydroxyl functional groups, and its super-hydrophobic π-π interactions. GO shows many useful properties including hydrophilicity, antifouling properties,[107] and high surface activity.[108] However, despite these advantageous properties, GO sheets suffer from two major drawbacks, namely, weak structural stability in the wet state and low permeation flux.[109] It had been observed that the incorporation of a layer of GO sheets upon CNF membranes by sequential water filtration improves the performance of the membrane.[48] These membranes also exhibited greater than 90% rejection of dyes, irrespective of

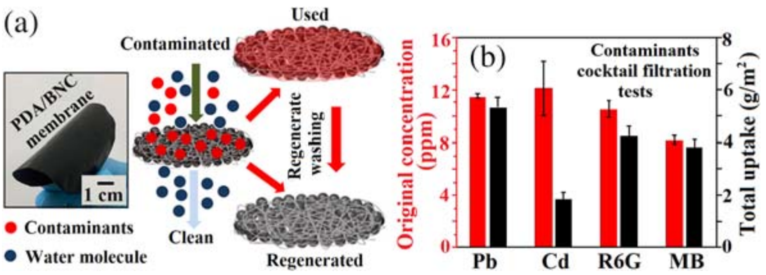

FIGURE 4 (A) Reusable and scalable BNC/PDA filtration membrane and (B) original concentration and the total amount of contaminants adsorbed by the BNC/PDA membrane[46]

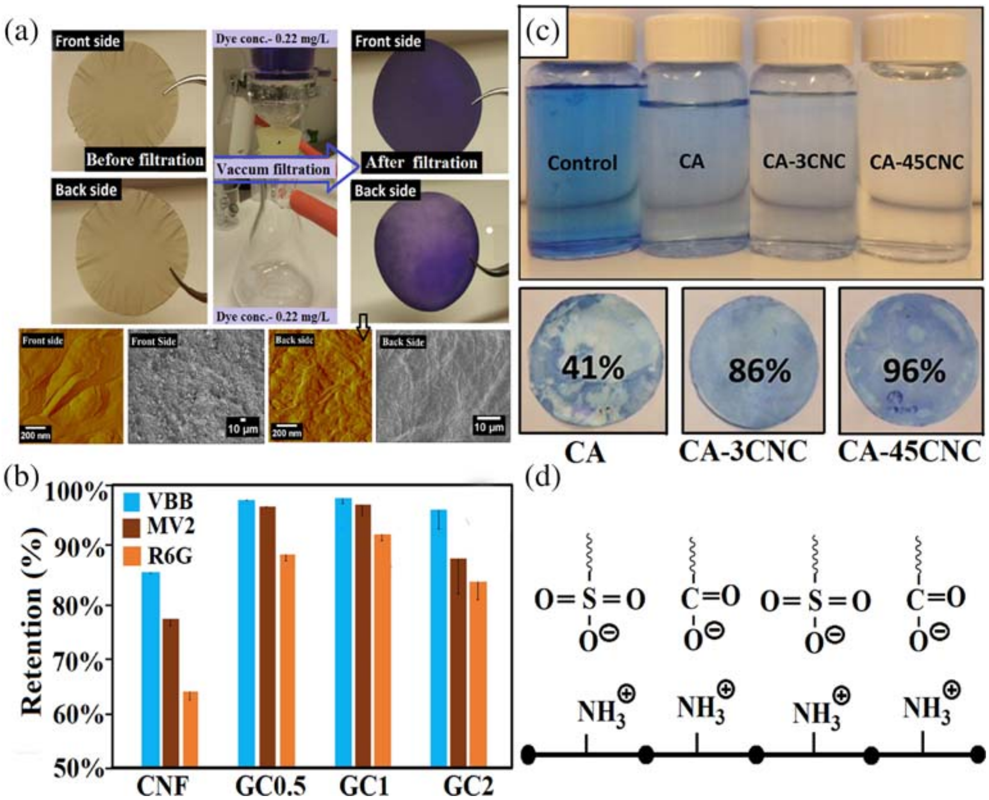

FIGURE 5 (A) Before and after photographs and micrographs GO-CNF1 membranes post dye filtration. (B) Graph depicting the adsorption efficiency of membranes having different Graphene concentration (GC0.5, GC1, GC2—CNF membranes with 0.5, 1 and 2 wt% concentration of graphene).[48] (C) Results after cross filtration of dyes using electrospun membranes and electrospun membranes coated with CNC (CA—Cellulose acetate fiber mats; CA-3CNC—CA loaded with 3 wt% CNC, CA-45CNC—CA loaded with 45 wt% of CNC).[121] (D) The mechanism of adsorption of dissolved colored compounds resulting from the electrostatic interaction between cationic and anionic groups [122]

the charge. It had also been observed that using a thin layer of GO as a functional layer on CNF could provide synergistic membranes mainly because of strong interactions between the two due to the formation of inclusive networks of inter- and intra-molecular hydrogen bonds.[110,111] Double layered CNF-GO membranes implementing an ultrathin GO layer of CNF membrane were prepared by Liu et al. without using any chemical crosslinker.[48] They used Exilva, a commercial-grade CNF, for fabricating the membrane, studied the impacts of the GO layer on its properties such as dye rejection behavior, mechanical properties, and surface characteristics. The study was carried out using VictoriaBlue 2B (VBB), Rhodamine 6 G (R6G), and MethylViolet 2B (MV2) as pollutants. They hence successfully exhibited the fabrication of a stable GO barrier layer onto a CNF membrane without using a chemical crosslinker. Figure 5A shows the photographs and micrographs of the membranes, before and after the vacuum filtration process. GO-CNF1 membrane exhibited the highest dye adsorption, with uniform distribution of GO (0.2 g/m^2). The membrane adsorbed 92.3%, 97.6%, and 98.8% of R6G, MV2, and VBB, respectively as can also be seen in Figure 5B. This membrane with around 100 nm thickness GO layer displayed higher water and dye permeation than monolayered GO membranes. The high rejection rate, structural stability, high flux, and easy preparatory method makes the GO-CNF membranes a worthy contender for membranes currently adopted for water remediation, food industry, and biomedicine.[112] From the tests conducted by Valencia et al.,[113] the results showed that the enhanced wet mechanical properties arose from the prevention of expansion of the CNF network during water filtration due to the incorporation of a GO layer, which also appeared to protect the cellulose network from variations in the structure during re-drying hence preventing dramatic collapse as occurs in a pristine CNF membrane.

Studies have revealed that NC either with additional surface functionalization such as enzymatic phosphorylation, TEMPO oxidation, cationization, and so forth or even in its native form, displays considerable adsorption efficiency in case of dyes, nitrates, humic acid, metal ions, and so forth from industrial effluents.[33,34,114–117] However, a challenge faced is the fabrication of a membrane with controlled pore size and structure, as well as the layering of several membranes over one another without compromising the membrane functionality, flux, rejection capacity, and the eco-friendly properties of NC. Electrospinning is gaining popularity as the route for the production of membranes with high surface area and an open interconnected porous structure.[118,119] Electrospun membranes have been used for adsorbing pollutants in water, at the micron scale. Goetz et al.[120] had previously shown that using chitin nanocrystals, biobased and standalone electrospun membranes based on cellulose acetate, having adequate mechanical properties, water flux, as well as anti-fouling performance, can be obtained. Goetz et al.[121] aimed at demonstrating an effective method to develop renewable membranes for water filtration, having high flux. They incorporated CNCs onto an electrospun mesh template prepared from cellulose acetate. The as-prepared membranes showed commendable antifouling properties, good mechanical stability, and were successful in rejecting contaminant particles ranging from 0.5 to 2.0 μm in size. The functionalization of cellulose acetate membranes using CNCs resulted in a highly efficient surface treatment approach for the preparation of biobased membranes that combine adsorption, size exclusion, and hydrophilicity (Figure 5C). The water flux was found to be very high (22,000 L m^{-2} h^{-1}) but decreased as the nanocrystal network became more continuous at a higher concentration. They also indicated increased hydrophilicity, as well as potential anti fouling towards hydrophobic entities. Its pore structure also showed moderate rejection of particles within 0.5–2.0 μm. The study demonstrated a simple and successful method to functionalize electrospun fibers using CNCs. The produced biobased membranes displayed improved mechanical properties, hydrophilicity, higher water flux, and pollutant rejection capability by size exclusion and adsorption.

Jonoobi et al.[49] prepared and characterized a composite membrane based on amine-functionalized CNCs and polyethersulfone (PES). They also studied its potential in chemical oxygen demand reduction and color removal from wastewater, a by-product of industrial licorice processing (Figure 5D). CNCs were modified using APTES. As discussed earlier, APTES increases the number of active sites in the composite, which are responsible for the elimination of impurities in water. The pristine PES membrane had the highest water contact angle (WCA) of 72.6°, showing a more hydrophobic nature. Whereas the WCAs of PES/0.1 wt% MCNC, PES/0.5 wt% MCNC, and PES/1 wt% MCNC declined, which indicated the hydrophilic nature of nanocomposite membrane surfaces. This could be attributed to the hydroxyl groups of amines, which exhibit good hydrophilicity, and CNC groups of APTES-modified CNC. The results obtained by them helped conclude that using APTES for the surface modification of CNC could offer a simple process, with no requirement of any solvent, by the prepared mixed matrix nanocomposite aimed at improving the color adsorption mechanism for industrial wastewater from licorice processing.

2.3 | Removal of salts

Polyamide based thin-film composites also showed promise for the filtration of water. Various strategies have been employed in the past few decades for improving the performance of polyamide-based thin film composite (TFC) NF membranes (NFM), such as the alteration of the reaction monopolymers,[123] the tuning of chemical and physical structures of the porous substrates,[124] the incorporation of nanomaterials into the porous substrate or polyamide film,[125–133] as well as the introduction of a layer in-between the polyamide film and the substrate. Of these, the incorporation of nanomaterials into membranes is more popular for modifying the permeation properties of the polyamide selective layers. Thin-film nanocomposite NFMs, or TFN NFMs, were used, which resulted in enhanced antifouling, chlorine, and antibacterial resistance attributed to the nanofillers. Figure 6A depicts the variation of the filtration performance of TFN NFMs varying with pressures. Various organic and inorganic nanomaterials have been developed but their preparation processes, unfortunately, tended to be complicated. Hence, an organic nanomaterial with a simple production process should have been found which also had superior stability for long term applications as well as good compatibility for the polyamide layer. One possible candidate is CNC which is very abundant and can be produced with ease by acid hydrolysis of cellulose.[134–140] It also displayed good antifouling properties when introduced into reverse osmosis membranes.[141,142] Huang et al.[50] observed the effects of CNCs on the properties of TFN NFMs. The membrane would become rougher and more hydrophilic, leading to higher performance than plain TFN NFMs. With an increase in CNCs, the water flux increased significantly by about 35% (increased to 106.9 from 78.9 L m^{-2} h^{-1}). The rejection of Na_2SO_4 remained above 98%, and that to $MgSO_4$ was above 96%, demonstrating good compatibility

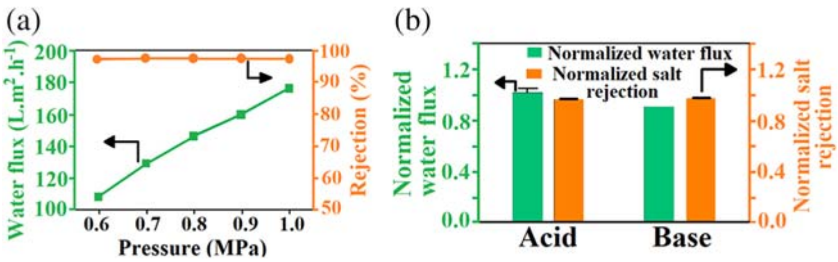

FIGURE 6 (A) Variation of adsorption through TFN-50 NFMs with pressures; (B) normalized salt rejection and water flux of the TFN-50 NFMs after alkaline (pH = 13) and acidic (pH = 1) treatments[50]

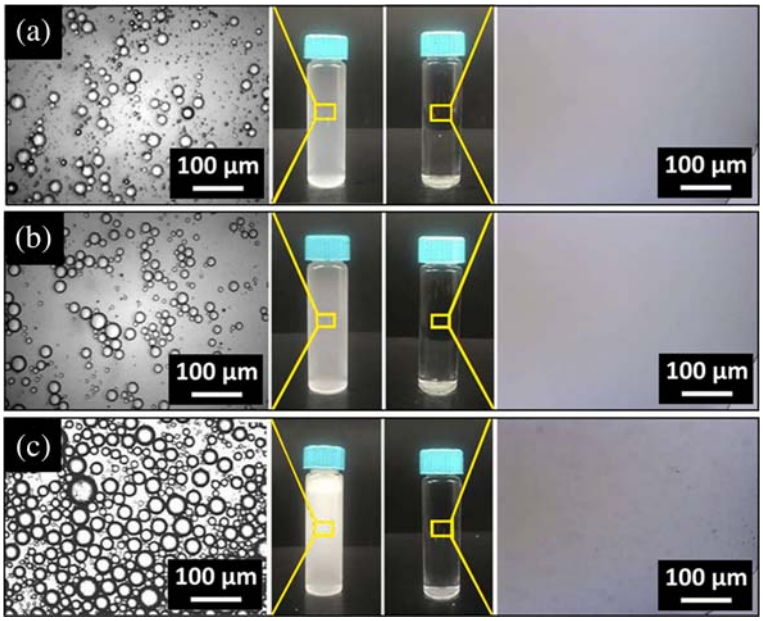

FIGURE 7 Microscopic images and photographs of (A) hexane, (B) petroleum ether, (C) Soybean oil, before and after separation[152]

between CNCs and polyamide matrix. It also displayed anti-chlorine property. Even after exposing the film to NaClO solution, it maintained a stable water permeation rate. Their study provides eco-friendly nanomaterials that are amazing nanofillers for the fabrication of TFN NFMs with enhanced water flux, without changing its salt rejection capabilities, which have been depicted in Figure 6B.

2.4 | Removal of oils

There has been a rapid growth in the petrochemical industry, resulting from the increasing consumption of petroleum resources. Dumping of oily waste products, into water bodies, or in particular, the transportation of crude oil-based products has frequently led to oil spills. Oil spills are a potential threat to both the marine ecosystem as well as human health. Membrane filtration is considered as a suitable method for separating oil/water emulsions.[14–16,18,143] However, commercial filter papers fail to separate oil/water emulsions due to large pore size and limited wettability. Tunicate CNCs (TCNCs) are isolated from the mantle of tunicate, a marine animal, hence they have excellent reinforcing ability and high aspect ratio.[144–146] TCNCs have been used as a 1D nanofiller for reinforcement of polymeric materials.[147–149] Membranes constituting of TCNCs obtained from the assembly of TCNCs or the co-assembly of TCNCs and various nanomaterials, display superhydrophilic, as well as superoleophobic surface owing to the intrinsic architecture and chemical composition of TCNCs.[150,151] In the study conducted by Huang et al.,[152] filter papers were modified using two strategies, by the physical method and chemical method. Their wettability and separation performances against various oil/water mixtures were measured, along with their stability in various conditions. The Oil Contact Angles (OCA) under the water, of both physically and chemically modified filter papers, decreased to ~148° from ~157°, with no significant change in the morphology of TCNC-filter papers even after undergoing 25 abrasion cycles. The underwater OCA of the filter papers remained high (about ~151°) after 20 cycles, indicating their superoleophobicity under the water. The results showed (Figure 7) that these modified filter papers showed super-hydrophilicity, nanoporous structure, and superoleophobicity, thus helping in the efficient separation of oil/water mixtures and emulsion. It also showed excellent UV resistance, tolerability of harsh environment, and anti-abrasion ability. Among the two methods of preparation, the membrane prepared by chemical modification demonstrated better anti-abrasion stability and higher efficiency in oil/water mixture separation.

3 | CONCLUSION

Good compatibility between the nanofiller and the matrix is indispensable considering the formation of interfacial channels, consequently increasing the water flux. Manufacturing such composites requires huge investment and complex processing methods. Also maintaining the structural integrity of the membrane without compromising to water flux and contaminant rejection efficiency is a major challenge. The research attempts highlighted in this review suggest that NC shows excellent compatibility with biopolymers and processing requirements for preparing NC based composites are minimal. Modified derivatives of NC allow the formation of active sites which show increased affinity towards the contaminant molecules via van der Waal's forces, electrostatic interaction and hydrogen bonding, and so forth. These active sites play a vital role in the adsorption of various contaminants including heavy metal ions, toxic dyes, oils, unwanted salts, and so forth. The processing methods used in the fabrication of these composites show negligible hindrance in its filtration properties while keeping their structural stability intact.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are openly available in repository name at http://doi.org/[doi], reference number [reference number].

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