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Nanocelluloses as sustainable membrane materials for separation and filtration technologies: Principles, opportunities, and challenges

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ABSTRACT

Membrane technology is of great interest in various environmental and industrial applications, where membranes are used to separate different mixtures of gas, solid-gas, liquid-gas, liquid-liquid, or liquid-solid. In this context, nanocellulose (NC) membranes can be produced with predefined properties for specific separation and filtration technologies. This review explains the use of nanocellulose membranes as a direct, effective, and sustainable way to solve environmental and industrial problems. The different types of nanocellulose (i.e., nanoparticles, nanocrystals, nanofibers) and their fabrication methods (i.e., mechanical, physical, chemical, mechanochemical, physicochemical, and biological) are discussed. In particular, the structural properties of nanocellulose membranes (i.e., mechanical strength, interactions with various fluids, biocompatibility, hydrophilicity, and biodegradability) are reviewed in relation to membrane performances. Advanced applications of nanocellulose membranes in reverse osmosis (RO), microfiltration (MF), nanofiltration (NF), and ultrafiltration (UF) are highlighted. The applications of nanocellulose membranes offer significant advantages as a key technology for air purification, gas separation, and water treatment, including suspended or soluble solids removal, desalination, or liquid removal using pervaporation membranes or electrically driven membranes. This review will cover the current state of research, future prospects, and challenges in commercializing nanocellulose membranes with respect to membrane applications.

1. Introduction

Membranes are widely used as selective barriers to solve environmental problems in industrial and municipal applications (Zuo et al., 2021). From water treatment, gas separation, and air filtration to food and biomedical applications, membranes play an important role in humans' quality of life. Various materials, including polymers, biopolymers, and inorganic substrates, are used to produce membranes for filtration and separation applications (Yang et al., 2019). In membrane contactors, the function of a membrane is to form an interface between two phases, not to control the flow rate of permeates through the membrane. The subject of membrane separation can be broadly defined to include not only the separation of gas mixtures, but also the two-phase separation of liquid phase components into a gas phase, a process known as pervaporation, and the separation of liquid phases as such. Separation of solids-rich solutions or concentrates from liquids or lean solutions is also possible with membranes. There are different types of separation membranes available, including nanofiltration

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membranes, reverse osmosis membranes, ultrafiltration membranes, and electrodialysis membranes, each designed for specific separation purposes (Esfahani et al., 2019). Chemical treatment of membranes involves exposure to a chemical agent, solvent, coupling agent, vapor, surfactant, surface treatment, or other additives. Physical treatment processes for separation membranes include ultraviolet irradiation, plasma irradiation, and sputtering.

Among the numerous nanomaterials being considered for membrane fabrication, nanocellulose is a unique biopolymer that can be obtained in different morphologies depending on its origin and processing conditions. The nanocellulose market is expected to grow from $346 million in 2021 to $963 million in 2026, at a compound annual growth rate of 22.7 % (Strathmann, 2001). It is difficult to estimate the end-user price of nanocellulose products, but it is estimated to be around $7 to $12 per kg of dried material. Nancelluloses are novel membrane materials for the transport or separation of components, species, or substances through or from the membrane. Nanocellulose membrane technologies are relatively simple in concept and operation, flexible, and compatible with integrated systems in many environments (Sharma, Sharma, Lindström, & Hsiao, 2020). Nanocelluloses are a suitable alternative for harmful and toxic polymers in membrane production and show high efficiency in various applications. Several authors have reviewed the advances in nanocellulose membrane research and fabrication. For example, nanocellulose composites developed for membrane and filtration technologies showed high performance in air filtration and purification (Rojas-Taboada & García Betancourt, 2022). Considering that removal of hazardous pollutants is crucial, filters for aerosol particle removal were also fabricated using nanocellulose composites and their removal by electrostatic charging was investigated (Norrrahim et al., 2021). Other work has demonstrated the efficient pendulum effect of nanocellulose composites for the adsorption and separation of various compounds from liquids. More recently, efforts have focused on membranes for water purification and treatment technologies (Tan, Ooi, & Leo, 2020a). Due to the physiochemical properties of nanocelluloses, these biopolymers are suitable materials for functionalization and surface modification to improve membrane performance. The commercial potential of nanocellulose membranes depends not only on the synthesis and modification processes but also on their commercial applications in desalination and membrane bioreactor systems.

In this review, we describe the use of nanocellulose membranes in various applications such as water purification, gas separation, air filtration, and other specific applications to solve environmental problems. The production methods of nanocellulose and combinations with other materials are explained for advanced environmental and industrial applications (gas mixtures, solid-gas mixtures, liquid-gas mixtures, liquid-liquid mixtures, and liquid-solid mixtures). Applications of nanocellulose membranes in different filtration and separation processes are discussed. Recent research on the preparation of nanocellulose membranes with tailored pore sizes, mechanical stability, rejection rate, adsorption capacity, and selectivity highly improves membrane properties. The performance of nanocellulose membranes combining high separation efficiency with antifouling properties is highlighted for overcoming large-scale production challenges. A technical evaluation of nanocelluloses will help to better assess their potential and, in particular, to think about their intrinsic properties and their translation into high-performance membranes.

2. Nanocellulose in membrane technology

Membrane technology has been on the rise since 1960 and is used in various industries such as water treatment, pharmaceuticals, food, biotechnology, dairy, and petrochemicals (X. Tan & Rodrigue, 2019b). Flexibility, easy scalability, cost efficiency, environmental friendliness, and low energy consumption make membrane technology an advanced field of research (Karim, Mathew, Grahn, Mouzon, & Oksman, 2014). The driving force for distinguishing different membrane-based processes depends on parameters such as pressure, concentration, temperature, or electrical potential gradient (Mohanty & Purkait, 2011). In particular, processes in gas separation, pervaporation, and evaporation membranes rely on the solution-diffusion mechanism in which molecules are transported along a thin, dense, and permeable selective membrane. The performance of membranes can be evaluated based on their productivity (i.e., influenced by critical parameters such as flow rate and permeability) and selectivity (i.e., separation factor) (Hokkanen, Repo, Bhatnagar, Tang, & Sillanpää, 2014). Overall, the history of nanocellulose in membrane technology is relatively short, but the material has already shown great potential for various applications. With continued research and development, we can expect to see new and innovative uses for nanocellulose in membrane technology in the future. The parallel development of membrane technologies and cellulosic materials as efficient filtration membranes is historically outlined in Fig. 1a.

Nanocelluloses consist of a family of cellulose materials with different nanoscale morphologies depending on the processing conditions. The materials and techniques used to prepare nanocellulose membranes determine their structural properties and subsequently the efficiency of the membrane in terms of transport properties. Fig. 1b provides an overview of recent publications on nanocellulose, including the different types of nanocellulose. Nanocellulose materials are produced from natural sources such as wood, plants, bacteria, or algae using different isolation methods, resulting in different morphologies. The production methods and morphologies of nanocellulose are described in detail and summarized in Table 1. The addition of organic and inorganic materials to cellulose leads to the production of nanohybrids and composites with controllable structures and mechanical properties, as described previously (Patel, Dutta, & Lim, 2019).

Advantages of nanocelluloses in membrane applications are related to their unique intrinsic properties, which are listed in Table 2. The nanocellulose membranes offer advantages over ceramic membranes, for example (Ahankari, George, Subhedar, & Kar, 2020; Zhu et al., 2015). Ceramic membranes are more expensive and difficult to manufacture but offer high stability and selectivity. Compared to ceramic membranes, nanocellulose membranes are characterised by high flexibility, light weight, low investment cost and energy consumption, environmental friendliness and non-toxic raw materials, simple manufacturing method, reusability and recyclability, and easy handling. Therefore, the incorporation of nanocellulose in membrane production can increase the pore size and porosity of the membranes, resulting in high water flux and selectivity. However, the disadvantages of nanocellulose membranes include shorter service life, weak chemical resistance, low operating temperature, tendency to membrane fouling, and weak high-temperature resistance. Hybrid nanocellulose membranes overcome some of these problems and introduce additional functionalities with improved durability. Grafting low molecular weight polymers and attaching polar functional groups on the nanocellulose membranes could improve the surface affinity and reactivity of the nanocellulose membranes for effective removal of certain contaminants. The incorporation of nanoparticles imparts new functionalities to the nanocellulose membranes.

Pore size of Nanocellulose membranes is the most important property that determines the selectivity of the membrane. It can be controlled by adjusting the processing conditions, such as the degree of fibrillation, surface functionalization, the drying method, and adding other materials to the membrane matrix. Based on the pore size of Nanocellulose membranes and filters, they can be classified into microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) (Baker, 2000; Baker, 2012). The operating conditions of the different membranes are determined by the pore sizes and operating pressures. Membrane filtration is based on the solubility and diffusion gradient of the various solutes. For UF and MF, size exclusion filtration is achieved by sieving particles through the membrane pores: the MF membranes separate suspended particles, macromolecules, and bacteria, while the UF membranes separate oil, proteins, and viruses. UF

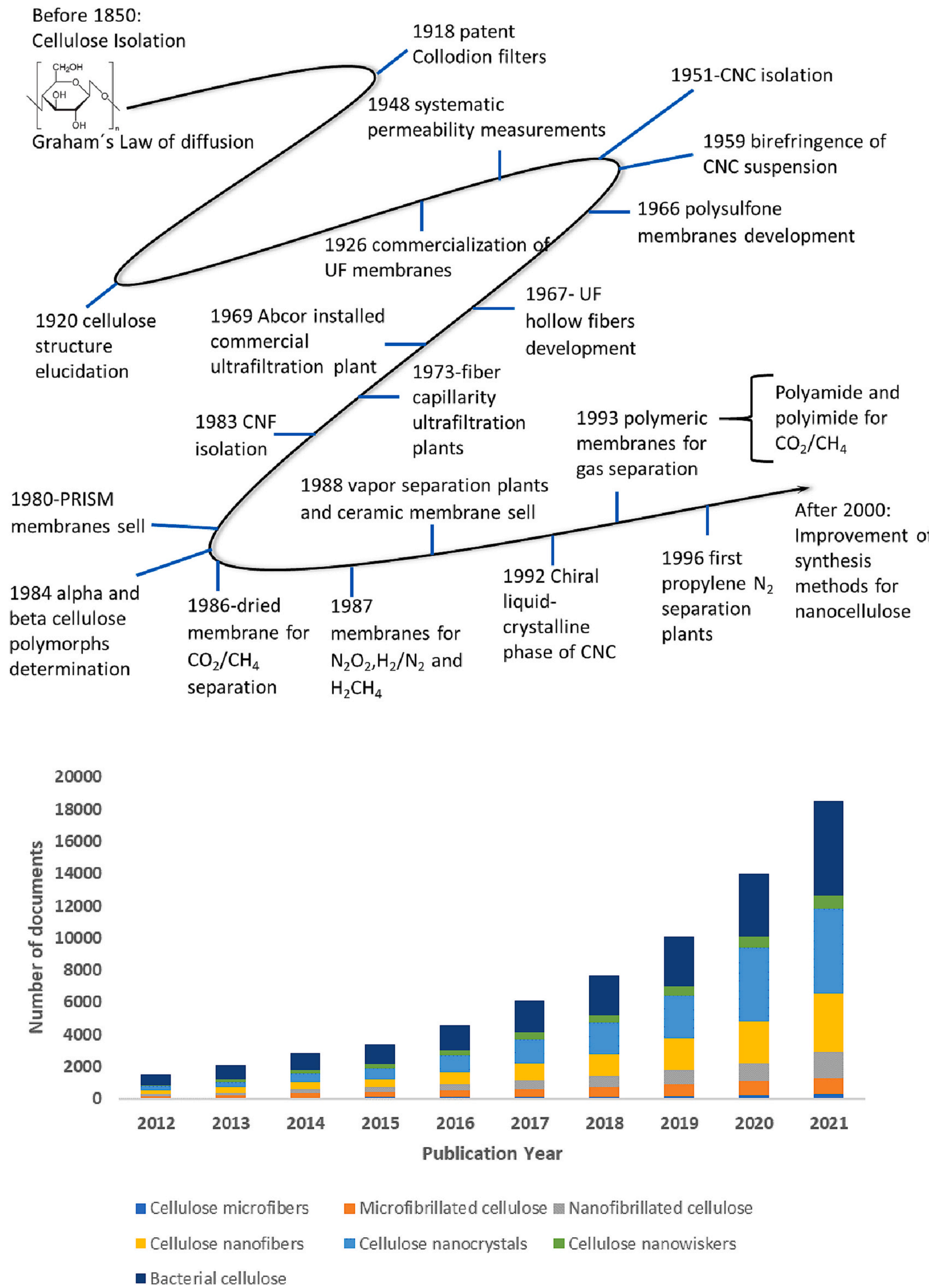

Fig. 1. Timeline for parallel development of nanocellulose separation membranes and data analysis of the number of publications in nanocellulose, performed on SciFinder using the terms "cellulose microfibers (CMFs), microfibrillated cellulose, nanofibrillated cellulose, cellulose nanofibers (CNFs), cellulose nanocrystals (CNCs), cellulose nanowiskers (CNWs), bacterial nanocellulose (BNC)".

is used for wastewater treatment, food processing, protein separation, and genetic engineering (Garcia et al., 2020). NF membranes separate particles at the molecular level and are generally used to separate divalent ions and various salts from water streams. The RO membranes have a very dense structure resembling a nanoporous structure as they are used to separate low molecular weight particles, such as organic molecules, and aqueous inorganic solids consisting of minerals, salts, and metal ions. The transport mechanism in RO membranes is diffusion through regions of free volume, with substances passing through the membrane and diffusing when pressure is applied. Desalination and drinking water production are the most common applications of RO membranes. Nanocellulose membranes are typically used for nanofiltration and reverse osmosis applications. It's important to note that pore size alone does not determine a membrane's selectivity or

Table 1
Production methods and preparation conditions for fabricating various nanocellulose types and derivatives.

Nanocellulose type	Production	Crys tallinity	Morphology	Aspect ratio	Ref.
Spherical cellulose nanoparticles (SCNPs)	Enzymatic or chemical pretreatment and hydrolysis (acidic and alkali treatment)	30–50 %	Spherical shape with 50–100 nm diameter	0.91–1.10	(Barhoum et al., 2020)
Nanocrystalline cellulose (NCC)	Chemical pretreatment (Alkali pretreatment, acid hydrolysis, oxidizing agent, organosolv, and ionic liquids)	54–88 %	Crystalline needle-shaped and rod-like cellulose particles with 54–88 % crystallinity	10–50	(Trache et al., 2020)
Cellulose nanofibers (CNFs)	Mechanical disintegration (Homogenizing, cryocrushing, microfluidization, grinding, and high-intensity ultrasonication)	68.5 %	Nanofibrils with a diameter of 10–100 nm, amorphous with crystalline sections	30–300	(Eyley & Thielemans, 2014; Habibi, 2014; Nguyen, Roddick, & Fan, 2012)
Bacterial Nanocellulose (BNC)	Biosynthesis (enzymatic pretreatment and bacterial synthesis)	75–80 %	Ribbon-like nanofibers with a diameter of 20–100 nm	50	(Barhoum et al., 2020)
Cellulose Nanoyarns (CNY)	Wet, dry, and melt spinning, or electrospinning of soluble cellulose derivatives such as cellulose acetate (CA) and hydroxypropyl cellulose (HPC).	50–60 %	Highly aligned fibril-like structures with a 100–1000 μm diameter	40–600	(Barhoum et al., 2020)
Cellulose nanohybrids	Melt extrusion and casting techniques using hydro-soluble, emulsion, and non-hydro-soluble systems	N/A	Different shapes with a few microns of length	N/A	(Barhoum et al., 2020)
Aerogels	Dissolution, gelation, and freeze-drying or supercritical drying. The Dissolution of cellulose followed by gelation and fabrication of the pore structure	N/A	A network structure containing fibers with 50–200 nm diameter, a foam-like structure with high porosity	N/A	(Barhoum et al., 2020)

performance. Other factors, such as membrane material, surface charge, and pore distribution, also play a significant role in determining the membrane's performance and suitability for a given application.

3. Fabrication methods of nanocellulose membrane

Nanocellulose membranes should have a combination of properties such as selectivity, permeability, chemical inertness, mechanical and thermal strength, and fouling resistance, as well as a simple and inexpensive fabrication method. The composition, processing methods, and nanocellulose loading can be optimized to achieve the desired performance, such as high selectivity, mechanical strength, chemical stability, and surface functionality (Tan & Rodrigue, 2019a). In particular, the mechanical strength and pore size of nanocellulose membranes strongly depend on the processing conditions. The choice of nanocellulose membrane components depends on the desired properties and target applications. Nanocellulose strengthens the mechanical strength of the polymer matrix and improves its thermal stability. By combining nanocellulose with ceramic materials, such as zeolites or metal oxides, composite membranes with unique properties can be produced. The nanocellulose provides mechanical reinforcement and improves the porosity and surface area of the ceramic matrix. These composite membranes can be used in applications such as molecular sieving, ion exchange, or catalysis. Graphene, a two-dimensional carbon material, can be combined with nanocellulose to form composite membranes with improved mechanical, electrical, and barrier properties. The combination of nanocellulose and graphene can result in membranes with improved water permeability, selectivity and conductivity.

Nanocellulose membranes are usually prepared by vacuum filtration of an aqueous nanocellulose dispersion on a filter paper substrate, followed by a drying process that also affects the filtration performance of the membranes. The more complex membrane structures are fabricated by thermal-induced phase separation (TIPS), non-solvent-induced phase separation (NIPS), vapor-induced phase separation (VIPS), melt spinning (MS), and cold stretching (CS), electrospinning, track etching, sintering, phase separation micro-molding, imprinting/soft molding, three-dimensional (3D) printing, and solvent casting (Tan & Rodrigue, 2019b). In addition, alternative methods such as physical blending, sol-gel, infiltration, film coating, solvent exchange, in-situ and interfacial polymerization, chemical atomic layer deposition, and layer-by-layer assembly and doping are used to fabricate nanocellulose composite membranes. These techniques can be used alone or in combination to fabricate nanocellulose membranes with specific properties, such as porosity, thickness, and surface charge. The choice of technique depends on the desired membrane properties and the intended application.

Vacuum filtration (Fig. 2) is following the methods of traditional papermaking and is used to produce the so-called nanopaper consisting of a stacked structure with nanocellulose layers. The nanocellulose suspension is drained through a filter membrane to form a wet cake with dense packing of the nanofibers forming a network with nanometer-scale pores. The resulting nanocellulose membrane obtained by vacuum filtration exhibits a layer of nanocellulose particles with a porous structure on the support membrane. The morphology of the membranes with the desired pore size and porosity can be controlled by the nanocellulose concentration, fiber length, and diameter (Mautner et al., 2015). The thickness and pore size of the membrane can be controlled by adjusting factors such as the concentration of the nanocellulose suspension, the filtration time, and the pore size of the support membrane. Vacuum filtration offers advantages such as simplicity, scalability, and the ability to produce membranes with high porosity. It is widely used in research laboratories and industrial settings for the fabrication of nanocellulose membranes for various applications. However, due to the formation of a dense layer with small pore sizes, dewatering and uniformity of the membranes during processing can be problematic and difficult to control.

Solution casting is a widely used method for fabricating nanocellulose membranes. It offers several advantages for the preparation of these membranes: (1) it controls the thickness and morphology; (2) it ensures the homogeneous dispersion of the nanocellulose in the solvent; (3) it allows the incorporation of additives such as polymers, nanoparticles or functional compounds; (4) it contributes to the wide range of applications in which nanocellulose membranes can be used. Typically, CNCs and/or CNFs are dispersed in a suitable solvent e.g. include water, organic solvents like dimethyl sulfoxide (DMSO), *N*-methylmorpholine-N-oxide (NMMO), or a mixture of solvents. After casting, the solvent is allowed to evaporate, either at room temperature or under controlled conditions like heating or exposure to a vacuum. The solvent evaporation leads to the formation of a solid nanocellulose membrane (Lizundia, Costa, Alves, & Lanceros-Méndez, 2020). Depending on the desired properties, the nanocellulose membrane may undergo additional treatments such as crosslinking, drying, or annealing to enhance its mechanical strength, stability, or other functional characteristics. It's worth noting that the specific details of the solution casting process, including the choice of solvent, additives, and casting method, can vary depending

Table 2
Relationship between physicochemical properties of nanocellulose and their efficiency in use as membrane material.

NC features	Effect on membrane efficiency	Reference
High surface area	The high surface area increases the adsorption capacity of NC compared to CMFBs.	(Patel et al., 2019)
High aspect ratio	The aspect ratio of CNCs (10–80) is generally smaller than that of nanofibers (up to 80–500), depending on the nanocellulose sources and treatment process. Favors the formation of percolated nanocrystals and entangled nanofibers networks held by strong hydrogen bonds, increasing the mechanical strength of the membranes	(Xu, Poggi, Resta, Baglioni, & Baglioni, 2020)
High mechanical stiffness	Highly crystalline forms (nanocrystals and nanofibers) are transparent and impermeable to gas and have very high tensile strength, up to 8 times that of steel.	(Barhoum et al., 2020)
High crystallinity	The high degree of crystallinity of nanocellulose (60–80 %) increases the chemical resistance of the adsorbent and reduces the solubility of cellulose even in highly polar solvents	(Tanpichai et al., 2012)
Susceptible to surface functionalization	The high content of negatively charged (hydroxyl) groups provides high activity for surface functionalization and increases the adsorption capacity of nanocellulose for a number of components.	(Habibi, 2014)
Stability in water	Negatively charged groups on nanocellulose lead to higher electrostatic repulsive forces between the surface layer and most model contaminants.	(Nguyen et al., 2012)
High surface tension	The high surface tension (the surface energy of nanocellulose is ~60 mJ.m^{-2}) of nanocellulose adsorbents by water improves the wetting properties and reduces biofouling	(Abdelmouleh et al., 2004)

on the specific nanocellulose material, desired membrane properties, and intended application. Optimization of these parameters is crucial to achieve the desired membrane performance. However, preparing membranes using this method takes a long time, and as a result, it is not suitable for industrial applications.

Freeze-drying is commonly employed for cellulose membranes prepared by solution casting because it helps preserve the structure and properties of the material. The low-temperature drying process minimizes the formation of structural defects and preserves the porous structure of the membrane. This is particularly advantageous for nanocellulose membranes, as it helps maintain their unique properties and high surface area. Freeze-drying and compaction were used to prepare CNC/Chitosan-UF membranes for water purification and dye removal. The latter was achieved by electrostatic interaction between negatively charged hydroxyl groups on the CNC surface and the positively charged dyes (Karim et al., 2014). Alternatively, the solution-casting nanocellulose membranes are made by a phase inversion process when immersed in a coagulation bath (Li et al., 2019). It involves the transformation of a homogeneous polymer solution into a porous membrane structure through a controlled change in solvent composition or temperature. After phase inversion, the resulting cellulose membrane is typically washed with a suitable solvent or water to remove any residual solvent or non-solvent. This step helps in the removal of impurities and ensures the stability and cleanliness of the membrane. This technique was used for the incorporation of CNC into cellulose acetate membranes, which were additionally kept in a water bath for several days to enhance the diffusion of the pore-forming polyvinylpyrrolidone into the aqueous medium.

Coating techniques can be used either to functionalize a given membrane material with an internal nanocellulose layer or deposit a nanocellulose layer on the membrane surface (Fig. 2). Here are some commonly used coating techniques for nanocellulose membranes: (1) Doctor blade coating is a fast and simple fabrication method in which nanocellulose is dissolved in a solvent and the homogeneous doping solution is poured onto a glass substrate using a doctor blade. (2) Dip coating is a traditional method for fabricating thin films of any polymer on cellulose material or vice versa. It involves immersing a substrate into a nanocellulose suspension or solution and then slowly withdrawing it, allowing a thin film of nanocellulose to form on the substrate. The immersion time can vary depending on the desired thickness of the membrane. The withdrawal speed influences the thickness and uniformity of the resulting membrane. The excess suspension drains off the substrate, leaving a thin nanocellulose film on its surface. Depending on the desired membrane properties, post-treatment steps such as cross-linking, annealing, or additional surface modification can be performed. (3) Spray coating is another technique where a nanocellulose suspension is atomized and sprayed onto a substrate to form a membrane. This method is suitable for large-scale production as it allows for high throughput and uniform deposition of nanocellulose on a large area. (4) Continuous casting or roll-to-roll processing involves the fabrication of nanocellulose membranes on continuous moving substrates. The nanocellulose solution or suspension is continuously coated onto the substrate and then subjected to subsequent processing steps, such as drying and post-treatment, in a continuous manner. This method is highly suitable for large-scale production due to its continuous and high-speed nature.

Melt blending (Fig. 3a) is a common method for fabricating nanocellulose composite membranes. A polymer matrix is chosen based on its compatibility with nanocellulose and desired membrane properties. Common thermoplastic polymers used in melt blending with nanocellulose include polyethylene, polylactic acid (PLA), polyvinyl alcohol (PVA), or others depending on the application requirements. After the melt blending process, the nanocellulose/polymer mixture can be formed into a membrane through various techniques such as extrusion, compression molding, or film casting. The specific method chosen depends on the desired membrane structure and properties (Gebald, Wurzbacher, Tingaut, Zimmermann, & Steinfeld, 2011). The specific processing parameters, such as temperature, shear rate, and mixing time, are optimized to ensure proper dispersion and distribution of nanocellulose within the polymer matrix. These parameters can affect the final properties of the nanocellulose membrane, such as its morphology, mechanical strength, and gas permeability (Hsu & Zhong, 2019). The resulting nanocellulose/polymer nanocomposite membranes can exhibit improved mechanical, thermal, and barrier properties, making them suitable for various applications such as filtration, separation, or packaging. However, using melt blending to produce nanocellulose membrane faces some problems such as the low dispersion of nanocellulose in the matrix, difficulties in mixing and changes of polymer viscosity, thermal sensitivity of nanocellulose, and preparation of uniform structures in the stretching process.

A popular chemical route for the synthesis of nanocellulose/polymer membranes involves toxic organic solvents. Therefore, many of the environmentally friendly methods described could only partially result in green synthesis. The ultimate goal is the fully green synthesis of nanocellulose membranes, where all raw materials, membrane production, post-treatment and other processes involved are “green”. In sol-gel polymerization, the polymer and nanocellulose precursors are mixed at the molecular level in a water-containing casting solution, and the

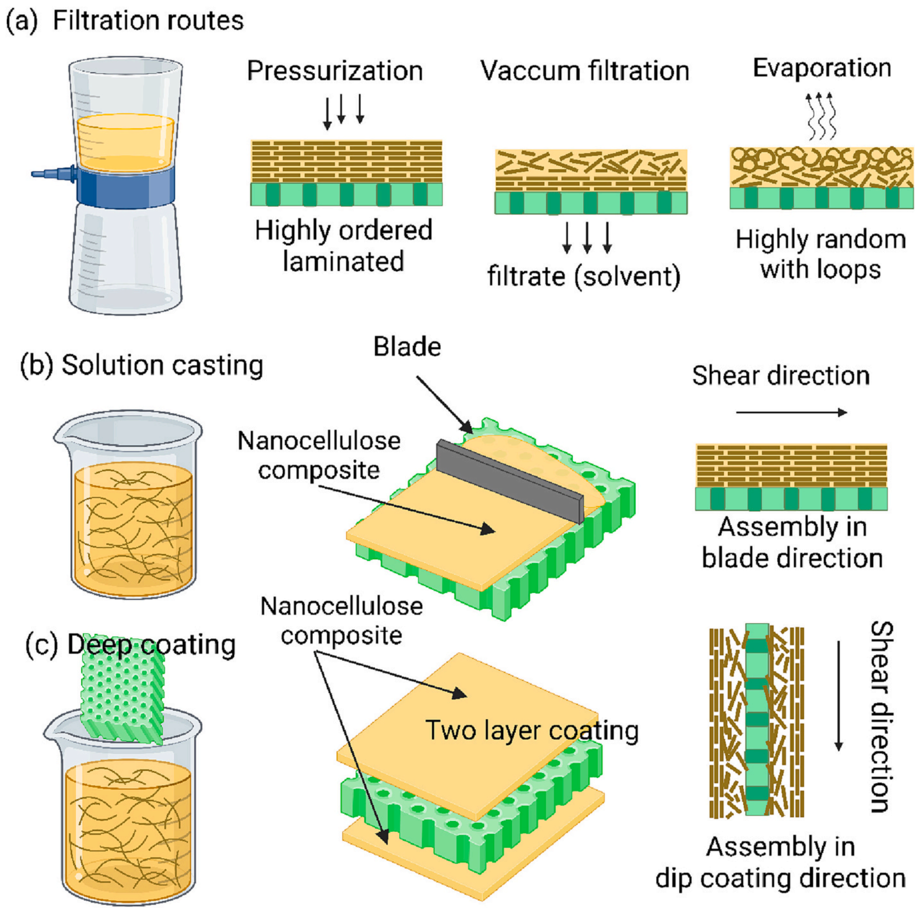

Fig. 2. Preparation of nanocellulose membranes: (a) dispersion of cellulose nanofibers/nanocrystals in a suitable solvent and wet filtration of the suspension; (b) dispersion of the nanocelluloses in a solution of polymer blends in the presence of a suitable solvent as a crosslinker and subsequent casting of the solution onto the polymer under adjusted conditions; (c) dip-coating a prepared membrane in a polymer solution containing nanocelluloses to deposit a surface layer on the membrane surface, to control the pore size and surface properties.

composite membrane is obtained by simultaneous sol-gel reaction and polymer precipitation. The membrane can be fabricated with a thin coating which shows excellent adhesion to the membrane surface. Thick coatings can also be produced as protective layers against corrosion. Simplicity, cost-effectiveness, high efficiency, high purity, and ease of control are among the advantages of this approach. However, contraction during processing, residual carbon groups, time-consumption, and toxicity of solvents hinder their application (Modan & Plăiașu, 2020).

In-situ and interfacial polymerization are two techniques used for the synthesis of polymeric membranes (Fig. 3c), which can also be used to fabricate nanocellulose composite membranes. (1) In-situ polymerization, which involves the simultaneous polymerization of monomers and formation of the membrane in a single step. It is typically carried out in a solution containing a cross-linking agent, a polymerization initiator, and a solvent. The nanocellulose is added to this solution, and the polymerization is initiated to form a polymeric matrix in which the nanocellulose is embedded. This technique allows for good control over the membrane morphology and properties. (2) Interfacial polymerization, in which the polymerization of monomers at the interface between two immiscible phases, such as an aqueous and organic phase. One phase contains a monomer, while the other contains a reactant that initiates the polymerization. The nanocellulose is typically added to one of the phases prior to the polymerization. This technique can lead to the formation of thin, dense membranes with good mechanical strength. Recently, nanocomposite membranes of CNC/polyamide were synthesized for RO and desalination by in-situ polymerization of *m*-phenylenediamine (MPD) and trimesoyl chloride (TMC) with different amounts of CNCs (Asempour, Emadzadeh, Matsuura, & Kruczek, 2018). Alternatively, interfacial polymerization could be performed with polyacrylonitrile (PAN) in green ionic liquids containing different nanocellulose additives to functionalize a PET membrane substrate (Wang, Ma, Chu, & Hsiao, 2017; Wang et al., 2014). It was found that interfacial polymerization with the aqueous phase over the organic phase (IP-R) gave a better filtration performance (Wang, Ma, Hsiao, & Chu, 2014; Turner & Liu, 2012).

Melt processing of nanocellulose membranes uses heat and pressure to shape and form the nanocellulose material into a membrane. Here are some common methods: (1) Extrusion: in this process, the mixture of nanocellulose and matrix material is heated to the melt state and then forced through a die to form the membrane (Fig. 3a). The extruded material is then rapidly cooled to solidify and retain its shape. (2) Mold pressing, in which the mixture of nanocellulose and matrix is heated to a molten state and placed between two heated plates or molds (Fig. 3b). Pressure is applied to compress the mixture and form a membrane. The membrane is then cooled to solidify. (3) Film casting: in this process, a solution or dispersion of the mixture of nanocellulose and matrix material is prepared in a solvent. The solution is applied to a substrate or casting surface and then dried or evaporated to remove the solvent, leaving a solid membrane. It is important to note that the specific parameters and conditions used in melt processing nanocellulose membranes can greatly affect the final membrane structure and properties. For example, melt processing techniques such as hot pressing (Fig. 3d) result in homogeneous and isotropic polymer/nanocellulose membranes, while the combination with melt stretching (Fig. 3e) can be used to create specific structures and align the nanocellulose along the stretching direction, resulting in anisotropic mechanical reinforcement (Fig. 3f).

Electrospinning is a simple method to produce nanofiber membranes, but its production capacity is limited. In this method, the polymer solution/melt is pulled under high-voltage electricity and the fibers are collected by a collector (Fig. 4a). The electrostatic forces are used to create a nonwoven fabric that acts as a membrane. The nanofibrous

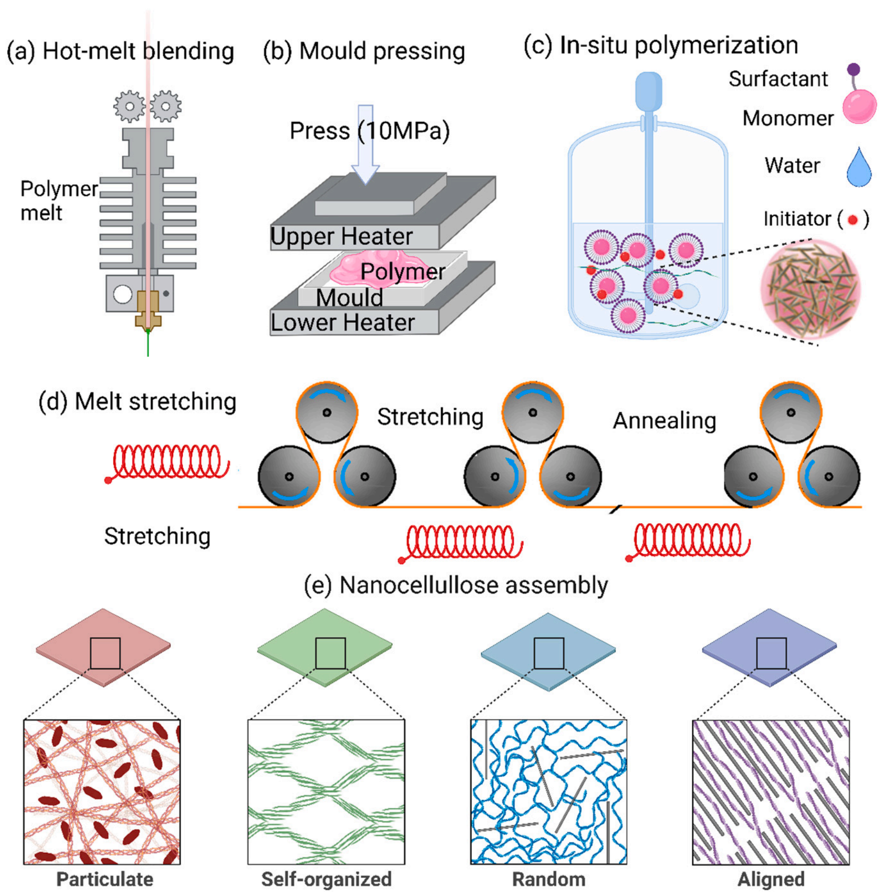

Fig. 3. Preparation of nanocellulose membranes: (a) hot melt extrusion of thermoplastic polymers with nanocelluloses; (b) compression molding of a thermoplastic or thermoset polymers loaded with nanocelluloses; (c) dispersion of nanocelluloses in polymer solution or in-situ polymerization of polymers in the presence of nanocellulose and subsequent preparation of membranes; (d) stretch molding of thermoplastic polymers loaded with nanocelluloses; (f) different orientations of nanocelluloses in the polymer matrix.

membrane can be collected on a rotating drum, flat plate, or other collector configurations depending on the desired membrane characteristics. After collection, the nanocellulose membrane may undergo additional treatments such as cross-linking, drying, or solvent removal to enhance its stability and mechanical properties. Various parameters such as the concentration of the solution, the type of nozzle, the spinning speed, the collector, and the environment can affect the morphology of the membrane. The resulting nanocellulose membrane obtained by electrospinning exhibits a fibrous structure with a high aspect ratio (length-to-diameter ratio) and a high surface area. The nanofibers in the membrane can be aligned or randomly oriented, depending on the electrospinning parameters and conditions. However, the solvent problem is the main challenge in this approach (Tan & Rodrigue, 2019b).

Compared to other methods, electrospinning enables the production of membranes with high pore interconnectivity and uniform pore size distribution. In addition, the membranes can be used with randomly oriented fibers in a core-shell fiber structure. Electrospun membranes are used for gas filtration, pervaporation, desalination, water treatment, and small particle removal (Mishra et al., 2019). The poly(vinyl alcohol)/nanocellulose membranes and porous films prepared by electrospinning provide good control over hydrogen bonding, crystallinity, and microscopic properties depending on the type and concentration of nanocellulose (Ji et al., 2021). The polycaprolactone/gelatin nanocellulose membranes were prepared after dissolving in suitable solvents to form a core or shell solution, while MgO nanoparticles were also added to the core solution under magnetic stirring (Peng et al., 2021). The electrospun nanocellulose composite membranes with incorporated carbon nanotubes and graphene oxide are particularly used for the treatment of dye wastewater (Moyo, Gumbi, De Kock, & Nxumalo, 2022). Electrospinning can be particularly used to fabricate high-performance composite membranes with core-shell nanofibers of CNC, carbon nanotubes, and poly(vinyl alcohol) (PVA) or poly(acrylic acid) (PAA) as crosslinking agents (Han et al., 2019). The membranes are used as flexible supercapacitor electrodes in combination with in-situ polymerization of polyaniline (PANI), which was used as a coating material, on the aligned electrospun nanofibers.

Centrifugal spinning is used as a high-throughput process to produce nanofibers, starting from a suitable solution placed in a central rotating spinning head (spinneret) (Fig. 4b). The fibers are stretched using a liquid jet and collected on the rotating collector, resulting in higher production rates, lower costs, and fewer safety issues compared to electrospinning. Centrifugal spinning is a simple and scalable process for producing porous fibers with oil sorption properties (Doan et al., 2019). The different morphologies for nanocellulose membranes can be obtained depending on the spinning conditions, including solid, mesoporous, hollow, or core-shell nanofibers (Barhoum et al., 2019). The highly porous composite fibers of cellulose and poly(vinylpyrrolidone) with micro- to nanoscale structures were obtained by tuning the parameters of centrifugal spinning and solvent evaporation, changing their performance in membrane applications (Hou, Li, Lu, & Yang, 2017). The lignin-amine/cellulose acetate nanofibers (LA/CA) were prepared by centrifugal spinning for membranes used for the adsorption of heavy metal ions (Xia et al., 2022), where the performance was optimized for membranes containing fibers with the highest lignin amine content (1.6 wt%) and the smallest average diameter (756 nm) as a function of spinning speed. The novel membranes with Janus cellulose acetate fibers were prepared by centrifugal spinning, resulting in membranes with combined superhydrophobic/hydrophilic properties for oil/water separation after gas-phase grafting of octamethylcyclotetrasiloxane (D4) onto the CA fiber membrane (Yu et al., 2021). Difficulty in pore size control has been a major challenge in electrospinning and centrifugal

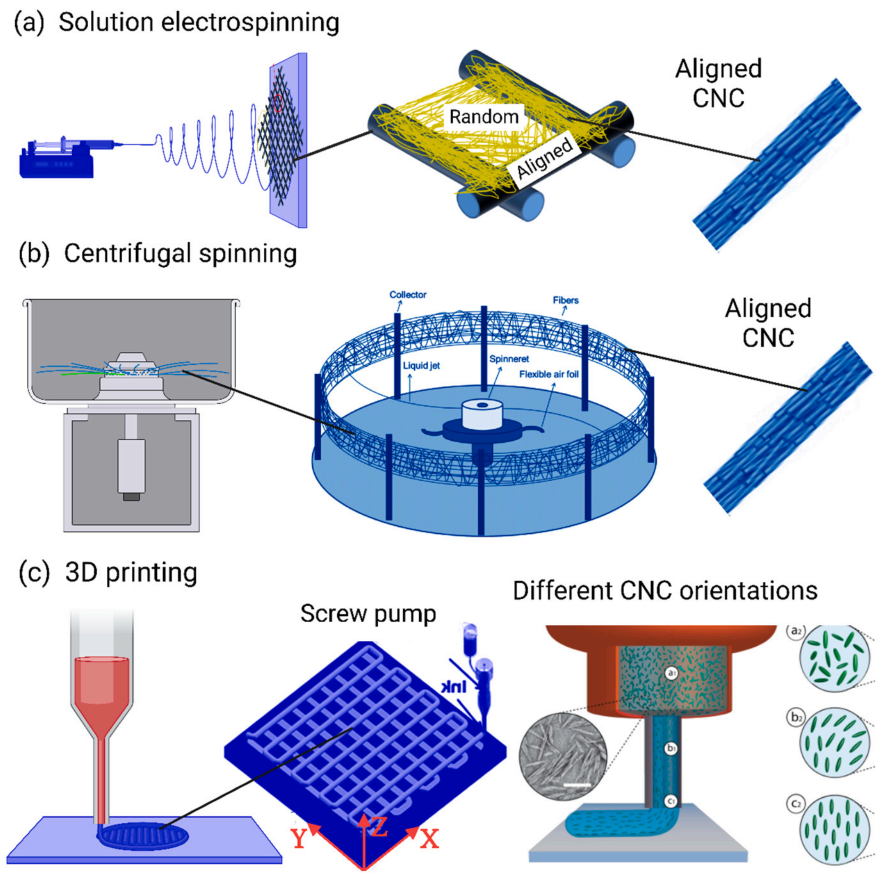

Fig. 4. Physical processes for the preparation of nanocellulose membranes: (a) electrospinning of polymer solutions or hotmelt containing nanocelluloses; (b) centrifugal electrospinning of polymer solutions or hotmelt; (c) 3D printing of polymer solutions containing nanocelluloses.

techniques. Fiber diameter determines the pore size, i.e. fibers of small diameters result in products with smaller average pore sizes. The decrease in the pore size is problematic as it causes lower infiltration. Additionally, these methods are limited to specific polymers and the use of solvents can also pose a threat to the environment (Dahlin, Kasper, & Mikos, 2011).

3D printing has recently been used for membrane fabrication, but resolution limitations are the main obstacle to direct 3D printing of membranes (Fig. 4c). Due to resolution limitations in the x and y axes, 3D printing becomes challenging where membranes of 0.1 to 1.0 μm pore size are required. Although the resultant membranes may show improved mechanical properties and high hydrophobicity, mechanical anisotropy is likely to happen as a result of the printing technology. Lower flexibility compared to conventional approaches and lack of suitable polymers also contribute to their limited use (Thiam, El Magri, Vanaei, & Vaudreuil, 2022). Nevertheless, this technology is very new for membrane fabrication and relatively unexplored for nanocellulose materials. Various technologies are used for 3D printing, such as material extrusion, powder bed fusion, vat polymerization, material jetting, binder jetting, and film lamination, with extrusion-based 3D printing (e.g., fused deposition modeling (FDM)) and filament free-form fabrication (FFF) are frequently used. In FFF, the molten thermoplastic material is deposited layer by layer until the 3D object is vertically formed. For FDM, selective laser sintering, stereolithography and poly-jet are popular 3D printing methods. The 3D-printed nanocellulose filters and membranes can be used in separation technology (as spacers and modules), water treatment and purification, solar steam generation, or environmental remediation, such as pollutant removal, filtration, and desalination (Finny, Popoola, & Andreescu, 2021). Accordingly, in a study conducted by Koh et al. (2019) a 3D-printed cellulose antifouling membrane was fabricated for oil/water separation applications using pure cellulose acetate/ethyl acetate solution as ink in direct ink writing. Under ambient conditions the solvent simply dries, resulting in an easily fabricated mesh architecture. In the following developments, all-cellulose membranes were fabricated by combining CNC on the surface of cellulose esters via large-scale 3D printing (Li et al., 2019), where the thickness, pore size, surface wettability, and water flux of all-cellulose membranes were controlled depending on the printing cycles of CNC inks. 3D printing of a CNF/graphite oxide (GO) mesh structure and integrating it as a layer in a composite membrane was designed for regulating water transfer through the membrane for steam generation (Li et al., 2017). The CNF/GO layer with a porous mesh-like structure transports water in its channels and conducts it upward to the adjacent porous CNT/GO layer. The porous and hydrophilic CNF/GO wall as the support can effectively draw water from the bottom due to capillary effects, creating continuous water transport pathways from the bottom to the top.

In the VIPS method, gas is used as a non-solvent that is exchanged with a solvent phase and causes membrane formation. Although it alters the morphology of the hollow fiber membranes, the use of VIPS is limited to commercial polymer membranes (Fig. 5a). Cellulose acetate separation membranes were prepared by VIPS with good control over microstructure as a result of composition ratios (solid content, acetone/*N*,*N*-dimethylacetamide ratio, glycerol/CA ratio) and membrane preparation conditions (evaporation time, evaporation temperature, and humidity) (Wang, Liu, Sheng, Zhu, & Yang, 2021; Wang et al., 2021). The high-performance membranes prepared through VIPS exhibit a homogeneous sponge-like pore structure and controllable surface morphology, which can potentially be used for bioseparation. The performance of nanocomposite membranes of polysulfone and CNF or CNC was compared by fabrication using immersion precipitation (IP) or vapor-induced phase separation (VIPS) (Daria, Fashandi, Zarrebini, &

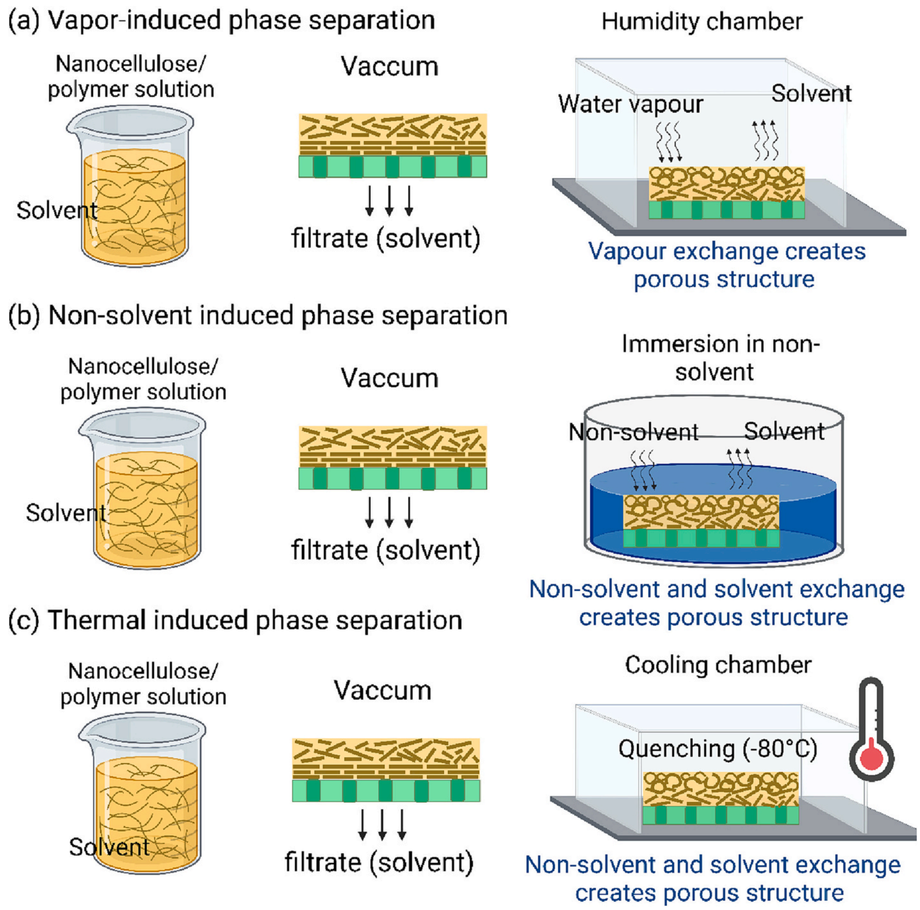

Fig. 5. Chemical routes for the preparation of nanocellulose membranes: (a) vapor-induced phase separation (VIPS); (b) non-solvent induced phase separation (NIPS), (c) thermally induced phase separation (TIPS).

Mohamadi, 2018), both of which indicated optimal concentrations of nanocellulose for the best water flux performance.

In the NIPS method, the membrane matrix and porosity are achieved by immersion of the polymer solution film in a solvent-free approach. In NIPS, the doped solution is the main first step in membrane fabrication, where the polymer and additive are dissolved in a suitable solvent and the obtained solution is poured onto the support layer (Fig. 5b). It is then precipitated into a coagulation bath with a non-solvent to produce the membrane. Due to the exchange between the solvent and the non-solvent in the coagulation bath, which is called liquid-liquid segregation, polymer-rich, and polymer-poor phases are formed during the phase inversion process. The polymer-rich phase forms the solid membrane matrix and the polymer-poor phase forms the membrane pores. The non-solvent acts as a pore-forming agent via diffusion of the solvent into the non-solvent (S Alibakhshi, Youssefi, Hosseini, & Zadhoush, 2019; Minelli et al., 2010; Ramasubramanian, Zhao, & Winston Ho, 2013). However, NIPS cannot be used alone to produce pure porous cellulose membranes. The porosity of membranes prepared by NIPS is relatively low due to the rapid exchange between solvent and non-solvent and the consequent precipitation of the polymer. The generation of porous structures from cellulose films during NIPS has been fundamentally studied while using ionic liquids (Wittmar, Koch, Prymak, & Ulbricht, 2020). The different viscosities of the cellulose solutions and the variable miscibility of the ionic liquids used with water strongly influence the porous structure of the film. The porous membranes for UF were prepared from cellulose acetate (CA) using the NIPS technique, where the porosity was controlled depending on the ratio of the casting solutions (e.g., acetone, and water) (Silva, Belmonte-Reche, & de Amorim, 2021). The nanoporous membranes were prepared after the regeneration of dissolved cotton pulp in NaOH/urea/water by NIPS using ethanol at different temperatures as a nonsolvent (Wang, Liu, et al., 2021). The limited solubility of ethanol in urea is an important parameter for controlling the nanoporosity of the cellulose membrane. The latter method is an alternative to the more expensive nanocellulose membranes and the use of cellulose derivatives. The method NIPS is alternatively used for the synthesis of a polysulfone network prepared from a pure polymer or a modified doping solution with nanocelluloses (Cruz-Tato et al., 2017). Thus, the metalized nanocellulose composites were prepared as candidates for the support layer of a forward osmotic membrane. The composite membranes of a mixed solution of polyvinylidene fluoride (PVDF) and cellulose were prepared via NIPS using *N*,*N*-dimethylacetamide (DMAc) and 1-butyl-3-methylimidazolium chloride as an ionic liquid solvent for PVDF and cellulose, respectively (Ma et al., 2017). The cellulose solution in the ionic liquid served as a porogenic product for the preparation of membranes with high hydrophilicity, wettability and high surface roughness.

The TIPS method is based on the segregation of a homogeneous polymer solution by temperature change and is a suitable method for various polymer membranes, especially for semi-crystalline polymers, while NIPS is suitable for pore size control by low polymer phases and surface modifications (Alibakhshi, Youssefi, Hosseini, & Zadhoush, 2021). This method is based on thermal energy (Fig. 5c). The polymer solution is heated above its melting temperature and then cooled until solidification and membrane formation. Phase separation micromolding is a microfabrication technique for various polymers in which a polymer solution is poured into a mold with a different structure, then phase separation occurs via TIPS or NIPS, and finally, the microstructure is released from the mold (Vogelaar, Barsema, van Rijn, Nijdam, &

Wessling, 2003). The process TIPS has been used to prepare FO membranes from cellulose triacetate together with dimethyl sulfone (DMSO2) as a crystallizable diluent and polyethylene glycol (PEG400) as an additive (Yu, Wu, Liang, Gu, & Xu, 2017). Pore size, porosity, water flux and mechanical properties of the membranes could be varied depending on the polymer concentration, the composition of the mixed diluent and the cooling condition. The hollow MF membranes of cellulose triacetate could be formed by TIPS, where the critical selection of suitable solvents was the main challenge and was solved by Hansen solubility parameter studies (Takao et al., 2022). The hollow cellulose triacetate membranes obtained by TIPS exhibited 3- to 5-fold higher mechanical strength compared to cellulose diacetate and cellulose propionate membranes. The cellulose membranes prepared from aqueous NaOH-urea solution and polyethylene glycol as a pore-forming agent was formed by NIPS and TIPS by immersing the casting solution into the coagulant under high temperatures while lowering the critical solution temperature (Hu, Niu, Chen, & Zhan, 2019). The high coagulation temperatures lead to rapid phase separation, resulting in membranes with large pores and loose pore structures. The latter morphologies are suitable for microfiltration and separation of oil/water nanoemulsions.

It is worth to mention herein that the NIPS is a versatile method and provides better control over pore size, but combining it with TIPS proves to be too expensive due to the additional energy required for heating. Overall, the parameters controlling phase inversion remain difficult to control due to challenges in kinetic and thermodynamic reactions in the presence of nanocellulose and need further optimization. A simplified route for the fabrication of hybrid nanocellulose membranes with multifunctional properties, optimization of processing conditions, and scaling up to economic costs are the main obstacles in the industrial breakthrough. All phase separation methods, namely VIPS, NIPS, and TIPS, cause safety hazards to the environment as the employed solvents are normally toxic. Additionally, these production methods are limited to a laboratory scale with a limited number of polymer configurations including dendrimers, diblock and triblock copolymers. Besides, the resultant fibers are rather short in comparison to some approaches (Ghalia & Dahman, 2016; Kulkarni & Rao, 2013; Zhao, Yildirimer, Lin, & Cui, 2016).

4. Air purification from airborne, pathogens, and volatile odors

Air filtration is the most widely used technology for removing particulate matter from an air stream because of its relative ease and flexibility of application (Zhao et al., 2019). The harm to public health caused by particulate matter (PM2.5) is the focus of attention worldwide. According to the Global Burden of Disease (GBD), PM2.5 is the fifth leading risk factor for death, causing 4.2 million deaths and 103.1 million disability-adjusted life-years (DALYs), representing 7.6 % of all global deaths and 4.2 % of global DALYs (Yang, Li, & Tang, 2020). Nanocellulose membranes for air filtration offer promising advantages such as high filtration efficiency, good air permeability, and potential antimicrobial properties. They have shown great potential for air filtration applications due to their unique properties, such as high surface area, mechanical strength, and biodegradability. The small pore size of nanocellulose membranes also allows for high selectivity in capturing fine particles and pollutants from the air. Although nanocellulose membranes provide effective filtration, they can also offer sufficient air permeability. The porous nature of the membranes allows air to pass through with minimal resistance, maintaining good airflow and minimizing pressure drop across the filtration system.

In air filtration applications, nanocellulose membranes can be used as a standalone filter or as a pre-filter in combination with other filters. They can capture a wide range of airborne particles, including PM2.5, PM10, bacteria, and viruses. Surface of nanocellulose membranes can be functionalized to increase their hydrophobicity or hydrophilicity, which can improve their efficiency in capturing certain types of particles. Nanocellulose membranes can be also functionalized with antimicrobial agents or surface-modified to exhibit antimicrobial properties. This feature helps inhibit the growth and proliferation of bacteria, viruses, and fungi on the membrane surface, reducing the risk of bioaerosol transmission. The thickness and pore size of the membrane can be controlled by adjusting the processing conditions. Ongoing research and development efforts aim to optimize their performance, durability, and scalability for various air filtration applications, including indoor air purifiers, automotive cabin filters, and personal protective equipment.

Hydrogen sulfide (H_2S) is a highly toxic air pollutant. A recent study was conducted to investigate the interest of MFCs treated with hydroxycarbonate apatite (HAP) and aminopropyltriethoxysilane (APS) in H_2S removal (Hokkanen, Repo, Bhatnagar, et al., 2014). The authors reported H_2S uptake values of 103.95 and 13.38 mg g^{-1} for APS/MFC and HAP/MFC membranes, respectively. An initial concentration of 80 mg l^{-1} H_2S solution was used for the experiments. Keshavarzi et al. (2015) described another method for odor removal using CNF/zeolite composite films with high adsorbent content. They found that these nanocomposites have a high affinity for volatile odors such as ethanethiol and propanediol with a charge of almost 90 wt%. Their results suggest that CNF zeolite films can drastically reduce the concentration of volatile odors below the detection limit of the human olfactory system (Keshavarzi et al., 2015). Recently, biodegradable composite air filters were fabricated from zeolite imidazolate framework-8@cellulose fibers (ZIF-8@CNF), Ag-based metal-organic frameworks@cellulose fibers (Ag-MOFs@CNF), and metal-organic frameworks@CNF (MOF-199@CNF) (Ma et al., 2018). The ZIF-8@CNF filter had filtration efficiencies of 98.4 % and 99.9 % for particle sizes of 0.3 μm and 0.5 μm, respectively, at a pressure drop of 134 Pa. Moreover, the nitrogen adsorption capacity of the composite membrane was about 20 times higher than that of pure cellulose filters. In addition, Ag-MOFs@CNF filters exhibited excellent antimicrobial activity against *Escherichia coli* (*E. coli*) with a prevention zone diameter of 20.8 mm. The composite filters prepared from MOF-199@CNF showed excellent filtration, antibacterial, and gas adsorption properties, and could be used in health care and air pollution removal. The release of organic ligands and metal ions can be considered as the antibacterial mechanism of MOF crystals, leading to bacterial cell membrane damage and DNA fragmentation (Ma et al., 2018).

5. Gas mixture separation membranes

Gas mixture separation involves separating different gases based on their molecular size or affinity for the membrane material. Nanocellulose membranes can be engineered to selectively separate a gas from a gas mixture, making them suitable for carbon capture and storage applications (Torstensen, Helberg, Deng, Gregersen, & Syverud, 2019). The performance of nanocellulose membranes for gas separation is typically evaluated based on several parameters, including permeability (flow rate), selectivity, and stability under operating conditions. Optimization of nanocellulose membrane properties, such as pore size distribution, surface modification, and membrane thickness, is essential for achieving the desired gas separation performance (Galiano et al., 2018). Optimization of pore size can be achieved by controlling the concentration of nanocellulose, modifying the surface chemistry, or adding additives that can create specific pore sizes or improve gas selectivity. Surface modifications can be used to improve the gas separation performance. For example, chemical grafting or coating it with nanoparticles can change the affinity of the membrane for certain gases, allowing selective gas permeation (Torstensen et al., 2019). However, it is worth noting that the development and optimization of nanocellulose membranes for gas separation is an ongoing area of research. Challenges such as membrane stability, permeability, and scalability need to be addressed to make nanocellulose membranes commercially viable for large-scale gas separation processes.

Separation using CNC and CNF-based membranes is considered one of the main options for CO_2 filtration using conventional technologies such as adsorption and absorption due to its great modularity, high cost, less or no release of chemicals, small footprint, and simplicity of the method. It should be noted that CNC and CNF differ in their crystallinity and length. The advantages of CNC are its homogeneous size in width and length with nanometric dimensions. The high tensile strength and aspect ratio of CNFs could be interesting for high-pressure gas filtration (Huang, Ou, Boving, Tyson, & Xing, 2009; B. Yu, Zhang, Shukla, Shukla, & Dorris, 2000). Surface functionalization of CNC and CNF can add new properties that modify functionality and uniformity of size distribution to produce membranes suitable for various gas filtration applications. Ongoing research and development in this field aim to further enhance the gas separation performance of nanocellulose membranes and explore their application in diverse industries, such as energy, environmental protection, and gas purification. The application of Nanocellulose membranes for gas mixture separation and CO_2/N_2 mixture separation at different relative humidity is depicted in Fig. 6. A systematic review on the feasibility of employing nanocellulose-based membranes for facilitated CO_2 separation from biogas was presented before (Nithin Mithra & Ahankari, 2022).

Owing to the very low gas permeability of nanocellulose membranes, they are usually used as gas barrier components and not for gas separation. Gas molecules can diffuse through the interconnected pore structure of the nanocellulose membrane, and separation can occur based on differences in molecular size, shape, or diffusivity. The type of nanocellulose used can impact the gas permeability of the membrane. For instance, CMFs, CNFs, and CNCs may exhibit different permeability properties due to their distinct structures and dimensions. Minelli et al. (2010) studied the transport properties of O_2 and H_2O in MFCs and discovered that CNF dry films have excellent barrier properties for O_2 and H_2O. However, when the water content in the membrane increases, the O_2 and H_2O permeability increases by 4–5 times. It is worth noting that CNF have specific structures that are more permeable than MFC films in the absence of water (Huang et al., 2009; Zhao et al., 2022). Nevertheless, the presence of hydroxyl groups in the molecular structure of fibrils increases the hydrophilicity of MFCs, promoting gas transfer due to the relationship between gas transport properties and water content (Minelli et al., 2010). Therefore, it was noticed that the MFCs exhibit unusual behavior compared to other cellulose-based materials because they are less affected by moisture. Other studies have shown that relative humidity increases CO_2 permeation and improves CO_2/CH_4 selectivity, where 1 % of crystalline nanocellulose at pH = 10 reported the maximum permeance of 0.29 m^3(STP)/(m^2-h-bar), with CO_2 selectivity over CH_4 being 43 (Jahan, Niazi, Hägg, & Gregersen, 2018).

CNC-based membranes show potential for various gas separation applications. CNCs act as reinforcing agents in nanocomposite membranes, thus utilizing the swelling mechanism at high concentrations and limiting moisture uptake (Jahan, Niazi, & Gregersen, 2018). Changing the CNC concentration in PVA membranes has shown a positive effect on membrane productivity at constant pH. The optimum CNC concentration in the PVA membrane was 1 wt%. At lower CNC amounts, the CO_2 permeation and CO_2/CH_4 selectivity of the membrane increased, while they decreased at concentrations higher than 1 wt%. This was attributed to the decrease in moisture uptake or the increase in crystallinity. A large increase in membrane thickness is also a drawback (Jahan, Niazi, & Gregersen, 2018). PVA/CNC and PVA/CNT membranes show similar separation performances with CO_2 permeance of 127.8 ±

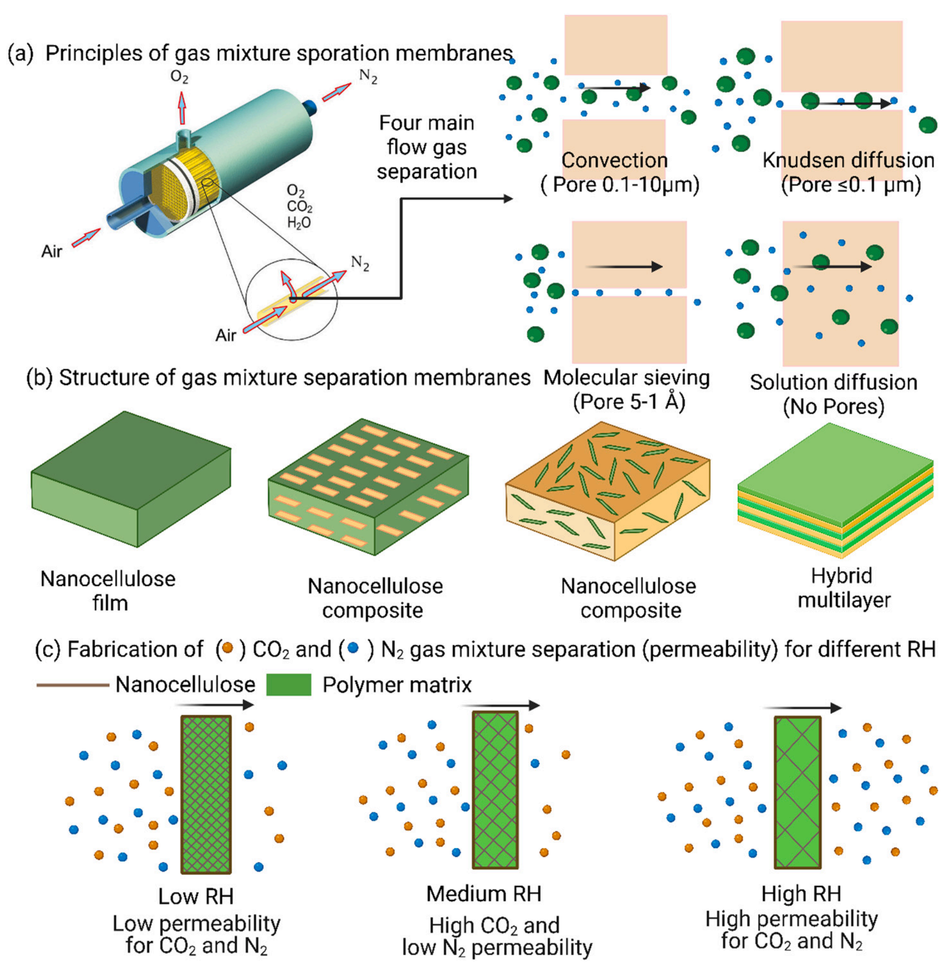

Fig. 6. Schematic presentation showing the application of nanocellulose in gas mixture separation membranes: (a) design and principles of gas diffusion membranes; (b) nanocellulose-based gas mixture separation membranes; (c) performance of CO_2/N_2 separation membrane at different relative humidity.

5.5 GPU and CO_2/N_2 separation factor of 39 ± 0.4 (Torstensen et al., 2019). Ongoing research aims to further enhance the gas separation performance of CNC membranes, explore new surface modification techniques, and investigate their application in diverse industrial sectors requiring efficient gas separation processes.

6. Water suspended matters removal

Removal of suspended solids occurs through several mechanisms (Jain et al., 2018), including: (1) size exclusion, i.e., physical exclusion of suspended solids above a certain size from passing through the membrane. The smaller the pore size, the more effective the membrane is at removing suspended solids. (2) Electrostatic attraction: the charge of the nanocellulose membrane can attract oppositely charged suspended solids and remove them from the liquid. (3) Adsorption, which can lead to the adsorption of suspended solids, such as bacteria and viruses. The performance of nanocellulose membranes in suspended matter removal depends on several factors, including the type and concentration of suspended particles, the pH of the solution, and the properties of the membrane, such as pore size, surface charge, and porosity. By tuning these factors, nanocellulose membranes can be optimized for specific applications, such as wastewater treatment, desalination, and food processing (Liu et al., 2021; Yuan et al., 2020). Fig. 7 illustrates the different mechanisms of NC membrane purification and wastewater recycling (Ahankari et al., 2020).

Microbial contamination of water by bacteria is a major health risk worldwide, especially in developing countries. For example, surface modification techniques can be used to alter the surface chemistry of the membrane and make it less favorable for microbial attachment and growth. This may include the use of antimicrobial coatings or surface charge modifications to prevent microbial attachment. The design of the membrane can also be optimized to reduce the potential for microbial attachment and growth. This may include the use of a narrower pore size distribution, a smooth surface, or a reduced thickness. Regular cleaning of the nanocellulose membrane can prevent the accumulation of organic material and the formation of biofilms. Cleaning protocols may include the use of detergents, solvents, or disinfectants. Prior to use, nanocellulose membranes can be sterilized using methods such as autoclaving or gamma irradiation to kill any microorganisms present. These methods can minimize the potential for microbial contamination in nanocellulose membranes, ensuring effective and safe water and liquid filtration.

CNFs and CNCs, with or without surface modification (e.g. cationization, tempo oxidation, and enzymatic phosphorylation), can adsorb pollutants from water through electrostatic interactions (Baker, 2012; Habibi, 2014; Tripathi, 2015). Virus and bacteria filtration and absorption of dyes are achieved using TEMPO-oxidized CNC micro fibrillation (Tan et al., 2020a; Tan, Ooi, & Leo, 2020b). Microparticles of 0.5 to 2.0 μm were removed using an acetate cellulose sheet, electrospun for wastewater treatment. Desirable antifouling properties, excellent mechanical stability, and suspended solid high rejection made the nanocellulose membrane a suitable choice for water treatment (Ahankari et al., 2020). The ideal membrane should have high flow at lower pressure, and thus require less energy while maintaining its high filtration efficiency (Ma, Burger, Hsiao, & Chu, 2012).

Various technologies and methods have been tested to improve the performance of nanocellulose membrane for suspended matters removal, especially chemical modifications. Interestingly, nanocellulose as a biopolymer is suitable for supporting MOFs to increase the tensile strength and porosity of the membrane. These membranes also have

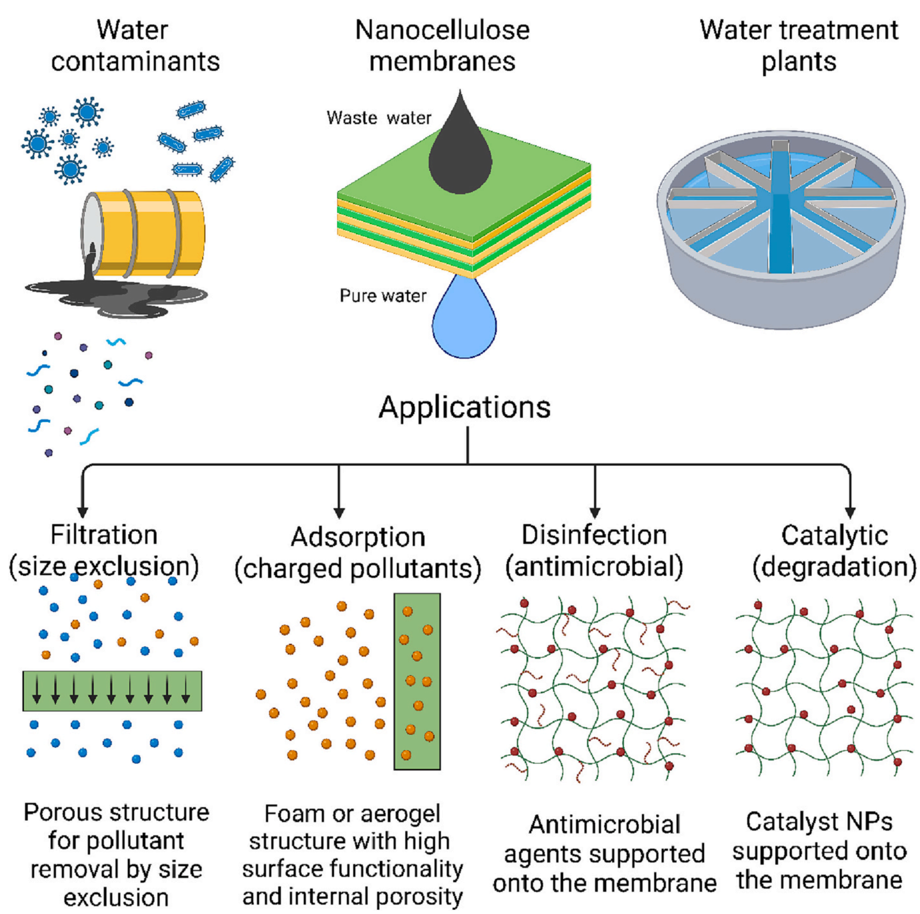

Fig. 7. Water/wastewater purification mechanisms include filtration, absorption, disinfection, and catalytic degradation.

larger pore sizes and improved water permeability for water purification (Cruz-Tato et al., 2017). TFC membranes can be achieved through the deposition of Ag and Pt NP on nanocellulose. They exhibit a finger-like porous morphology that contributes to the improvement of water flux and rejection of the membrane (Cruz-Tato et al., 2017). Grafted nanocellulose has also been used for wastewater treatment as it can trap pollutants in its functional groups (Bagheri & Julkapli, 2018). The water treatment cost is estimated to decrease by improving the hydrophilicity of the nanocellulose membranes by controlling the surface charges (e.g., carboxyl, carboxylate, amino, thiol, silanol groups, and nanoparticles which render surface charges and reactivity), or improving the mechanical strength of the membrane (Tan et al., 2020a, 2020b).

7. Membrane for water soluble matter removal

Nanocellulose membranes have shown a potential to remove water-soluble substances from various environmental matrices. The following are some of the potential applications of nanocellulose membranes for the removal of water-soluble substances: (1) Heavy metals such as lead, cadmium and mercury. (2) Organic pollutants such as dyes and pesticides. (3) Dissolved organic matter (DOM) responsible for color, taste, and odor problems. (4) Salts from water, such as in desalination processes. With continued research and development, we can expect to see new and innovative uses for these membranes in the removal of water-soluble matter. Water-soluble substances pollution is one of the most serious environmental problems (Karim et al., 2014). Many techniques have been used to remove water-soluble substances from wastewater, such as adsorption, membrane separation, chemical precipitation, electrochemical processes, ion exchange, flotation, and coagulation-flocculation.

Cellulose and its derivatives are particularly effective in the removal of environmental pollutants containing heavy metals (Ni^{2+}, Hg^{2+}, Cr^{6+}, Cu^{2+}, Cd^{2+}), organic pollutants, anionic and cationic dyes, and inorganic anionic contaminants (Carpenter, De Lannoy, & Wiesner, 2015). Nanocellulose adsorption membranes eliminate heavy metals by two main methods: chemical complexation and ion exchange, as shown in Fig. 8 (Mahfoudhi & Boufi, 2017). In ion exchange, metal ions are adsorbed and replaced by other ions associated with the adsorbent surface. In chemical complexation, the functional groups of the adsorbent surface interact with specific metal ions. In both mechanisms, stoichiometric rules limit the maximum absorption volume to half the amount of ionic surface dots. Therefore, surface modifications are essential to enhance or introduce complex ionizable ionic sites on the nanocellulose surface to absorb metal ions (Hokkanen, Repo, & Sillanpää, 2013). Amine and carboxyl sulfate groups are among the most widely used groups to improve adsorption capacity. These groups can be added during nanocellulose fabrication or by modifying the nanofiber surface.

Most studies on the use of nanocellulose for heavy metals removal have focused mainly on CNFs and rarely on CNCs. CNCs extracted from rice straw were evaluated as adsorbents for Pb^{2+}, Ni^{2+}, Cd^{2+}, Ag^{+}, Cu^{2+}, and Fe^{3+} with adsorption valence values of 9.7, 9.42, 8.55, 56, 20, and 6.5 mg g^{-1}, respectively (Kardam, Raj, Srivastava, & Srivastava, 2014). After surface functionalization with phosphate groups by enzymatic treatment, the cation sorption to CNCs increases to 117, 136, and 115 mg g^{-1} for Cu^{2+}, Ag^{2+}, and Fe^{3+}, respectively (about twice that of

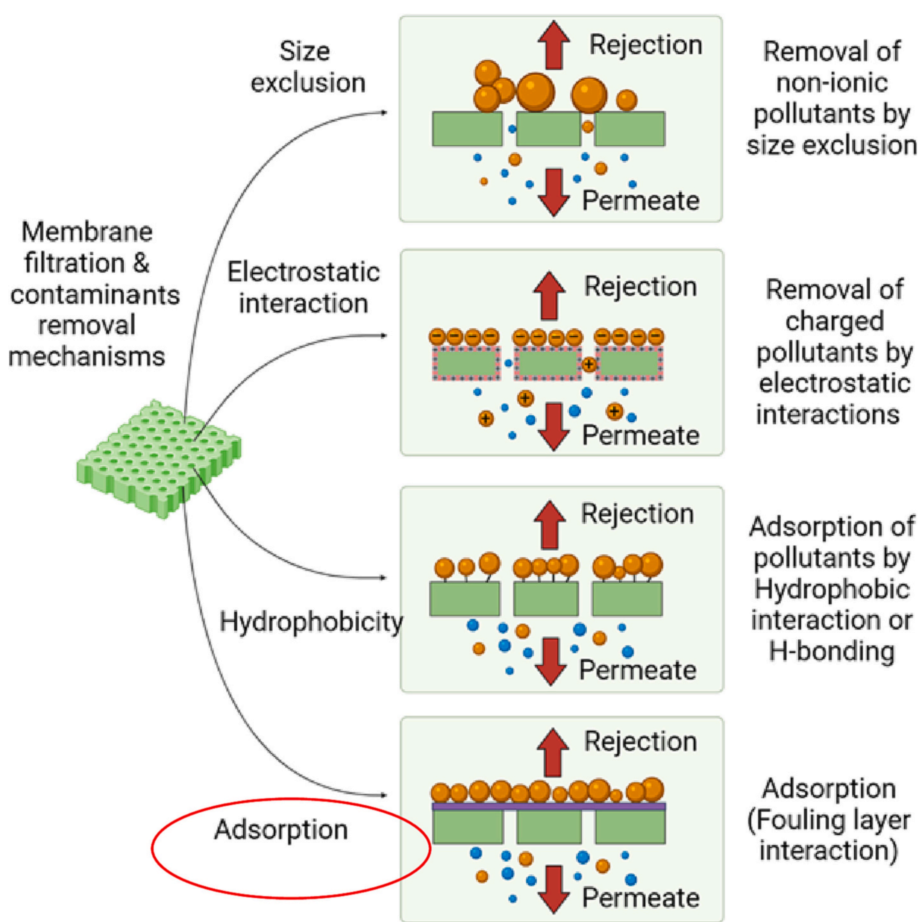

Fig. 8. (a) The mechanism of complexation and ion exchange in adsorption processes (Mahfoudhi & Boufi, 2017). Various methods for micropollutants removal: (b) size exclusion (c) adsorption (hydrophobic interaction), (d) electrostatic repulsion, and (e) adsorption (fouling layer interaction) (Das et al., 2017; Ojajuni, Saroj, & Cavalli, 2015).

untreated CNCs) (Liu et al., 2015). The elimination efficiency is favorable due to the specific surface area of these nanomaterials prepared with functional groups on the nanocellulose surface. Adsorption is better at neutral pH, and membrane regeneration can be successfully performed with nitric acid. Modification of the CNC surface with succinic anhydride improves the adsorption capacity of the membrane for Pb^{2+} and Cd^{2+} by 10 times compared to untreated CNC. The adsorbent was effectively reconstituted with saturated NaCl solution after two cycles without any loss of activity (Yu et al., 2013). Amination of BC with diethylenetriamine resulted in adsorption valence values of 40 and 50 mg g^{-1} for Pb^{2+} and Cu^{2+}, respectively, at pH ~ 5 in 2 h (Chaker & Boufi, 2015).

Aminated BC was successfully regenerated by washing with an acidic solution or EDTA to separate the adsorbed metal. In this work, ionic or ionizable groups such as COOH, SO_4^{2-}, and -R_3N^+ were applied to the surface of CNFs either before or after the preparation of fibrils. However, most of these groups are added as part of a chemical pretreatment process prior to CNF fabrication because they facilitate the fibrillation process by creating repulsive forces between the microfibrils that contribute to the weakening of their cohesion through hydrogen bonding (Chaker & Boufi, 2015). Carboxylation of CNF has also been evaluated as a method to introduce binding sites on the surface of nanofibrils (Galiano et al., 2018; *Nanocellulose-Based Products for Sustainable Applications-Recent Trends and Possibilities*, 2020). Such carboxyl groups increase the adsorption capacity more than twofold compared to unmodified BC: 20 mg g^{-1} (Cu^{2+}) and 65 mg g^{-1} (Pb^{2+}) versus 11 mg g^{-1} (Cu^{2+}) and 25 mg g^{-1} (Pb^{2+}) for pure BC (Chen et al., 2009). Adsorption is determined by the fraction of divalent metal ions with amino functions via complexation and ion exchange. The APS/CNF membrane can be regenerated by alkaline treatment (Hokkanen et al., 2014). CNFs modified with succinic anhydride were also used for the adsorption of Cd^{2+} and Pb^{2+} ions in wastewater, with maximum adsorption of 3 and 12 mmol g^{-1}, respectively, compared with 0.002 mmol g^{-1} for crude cellulose (Stephen et al., 2011). The modified nanofibers were regenerated to a neutral pH by washing with HNO_3 and repeated rinsing with distilled water.

A comparison of different studies showed that oxidized nanofibers performed better compared to many other sorbents from bio-based sources in terms of adsorption of heavy metal ions, such as copper adsorption using sugar beet pulp (0.33 mmol g^{-1}) (Reddad, Gerente, Andres, & Le Cloirec, 2002), aspen wood fibers (4 mg g^{-1}) (Huang et al., 2009), sawdust (1.79 mg g^{-1}) (B. Yu et al., 2000), wheat bran (51.5 mg g^{-1}) (Özer, Özer, & Özer, 2004), wheat (17.42 mg g^{-1}) (Aydın, Bulut, & Yerlikaya, 2008), jute fiber (8.4 mg g^{-1}) (Shukla & Pai, 2005), herbaceous peat (4.84 mg g^{-1}) (Gundogan, Acemioğlu, & Mehmet, 2004), bentonite clay (44.84 mg g^{-1}) (Bertagnolli, Kleinubing, & Silva, 2011), untreated and pretreated seaweed (6.12 mmol kg^{-1}), and kaolinite (20 mmol kg^{-1}) (Suraj, Iyer, & Lalithambika, 1998). Oxidized nanofibers provide similar or lower copper adsorption capacity than mercerized cellulose modified with succinic anhydride (139 mg g^{-1}). Another method for the separation of heavy metals is magnetic separation and purification as described by Ambashta and Sillanpää (2010). These authors fabricated magnetic nanocellulose using Fe_3O_4 nanoparticles with embedded BC spheres as hybrid nanocomposite adsorbents to adsorb Cr^{3+}, Pb^{2+}, and Mn^{2+}. The spherical Fe_3O_4/CNF nanocomposites showed adsorption capacities of 25, 65, and 33 mg g^{-1} for Cr^{3+}, Pb^{2+}, and Mn^{2+}, respectively. These BC amino magnet nanofiber nanocomposites showed higher adsorption capacity than As^{5+} and also compared to nanofibers (about 1.2 times higher) and iron oxide base oxide (2.4 times higher). The regeneration was carried out by alkaline treatment (Nata, Sureshkumar, & Lee, 2011).

As mentioned earlier, CNF-based membranes are increasingly used for water treatment. In particular, distillation membranes were developed to improve filtration by removing heavy metal ions. Accordingly, thermal crosslinking of thiol-modified CNFs with the electrospun nanofiber scaffold PAN resulted in the formation of super-filtration membranes composed of nanofibers adsorbing Pb^{2+} and Cr^{6+}. Such membranes could be regenerated up to three times without loss of adsorption capacity by rinsing in an EDTA solution (Yang et al., 2014). The grafting of many monomers is one of the golden steps to increase the adsorption capacity of CNFs by increasing the available ionic sites to which the metal can bind. Poly(methacrylic acid-*co*-maleic acid) grafted CNFs prepared using Fenton's reagent in water form a highly porous aerogel, which increases the availability of the adsorption surface and allows the adsorbent to be resorbed and reused multiple times (Maatar & Boufi, 2015). The aerogel with density in the range of 0.03–0.06 g cm^{-3} has a pore size range of 50 to 500 μm. The maximum adsorption capacities for Ni^{2+}, Pb^{2+}, Cd^{2+}, and Zn^{2+} were 138, 165, 135, and 117 mg g^{-1}, respectively, which were three times higher than those obtained with pure CNFs. At initial concentrations of <10 ppm, the adsorption rate for divalent metal ions was >95 %. It should be noted that the adsorbent can be regenerated without loss of adsorption capacity by simply washing it in EDTA solution.

8. Membranes for liquid-liquid separation

Nanocellulose membranes have shown promise for use in liquid-liquid separation applications due to their high porosity and unique surface properties. The following are some of the potential applications for liquid-liquid separation of nanocellulose membranes: (1) Emulsion separation such as oil-in-water or water-in-oil emulsions. The pore size and surface chemistry of the nanocellulose membrane can be tailored to optimize separation performance for specific emulsions. (2) Solvent recovery, for example, the recovery of organic solvents from industrial processes for reuse. Membrane pore size and surface chemistry can be optimized to selectively retain or pass specific solvents or solutes for efficient separation. (3) Separation and purification of pharmaceutical and biotechnology products, for example, suspended solids, colloids and fine particles cab be removed from liquids while allowing the desired liquid phase to pass through. (4) Used in membrane distillation processes where a liquid mixture is heated on one side of the membrane while maintaining a temperature gradient. The vapor generated on the hot side passes through the membrane, leaving behind the separated liquid components (Gupta, Dunderdale, England, & Hozumi, 2017).

Emerging cellulose-based aerogels have been used for oil absorption applications. The hydrophilic property of nanocellulose associated with their molecular structure is not suitable for oil absorbents (Jin et al., 2011). To solve this problem, a surface modification is required to transform the CNF-based aerogel into an extremely hydrophobic material that behaves like a hydrophobic reservoir for oil and hydrocarbons. As shown in Fig. 9, Zhang, Sèbe, Rentsch, Zimmermann, and Tingaut (2014) transformed a CNF aerogel into an effective reusable absorbent for oil by silylation with methyltrimethoxysilane. Porous sponges were prepared by freeze-drying CNF-based aqueous suspensions containing methyltrimethoxysilane as silylating agent. The silylated material showed oleophilic and hydrophobic properties, which could be used to selectively remove spilled dodecane on the water surface (with a maximum removal of 52 g g^1). These CNF aerogels exhibit high absorption capacities for a wide range of organic solvents and oils (up to 102 g^{-1}).

Tarrés et al. (2016) synthesized a hydrophobic CNF aerogel by freeze-drying and chemically grafting alkyl ketene dimer (AKD) onto the CNF surface by esterification at 110 °C for 10 min. The AKD-modified CNF aerogel was used as a reusable oil absorbent with exceptional absorption capacity (> 140 g g^{-1}). Feng, Nguyen, Fan, and Duong (2015) developed cellulose aerogels from paper waste using a Kymene cross-linker to improve the gelation process instead of alkali/urea. They treated freeze-dried CNFs with methyltrimethoxysilane (MTMS) to convert the aerogels into a hydrophobized material. The membrane containing 0.25 wt% cellulose aerogel exhibited the highest oil absorption capacity (95 g g^{-1} at 50 °C). The sorption capacity was affected by the viscosity of the oils tested, but was not dependent on the pH of the

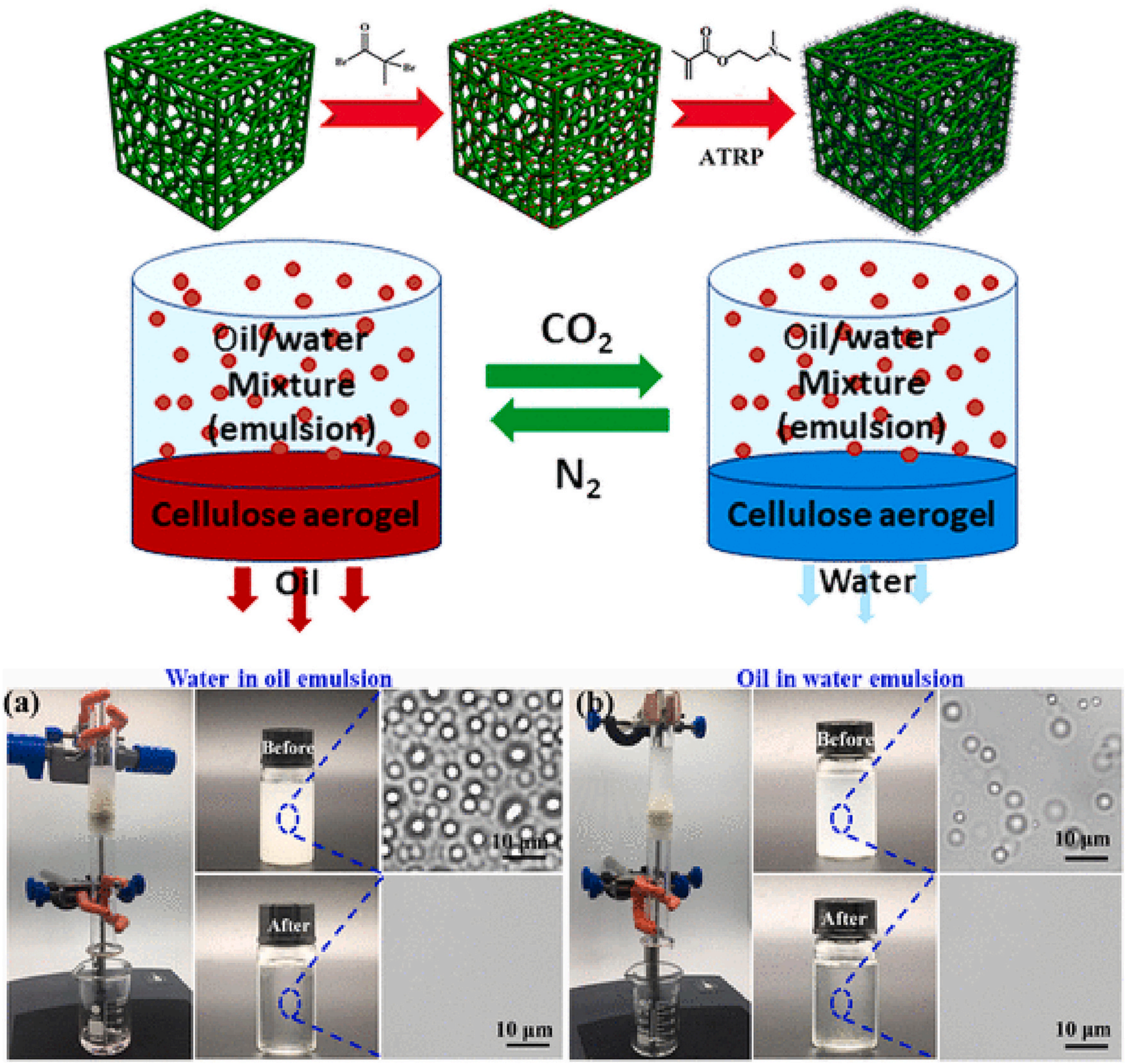

Fig. 9. CO_2-sensitive cellulose nanofiber aerogels for switchable oil-water separation. Light microscope images and photos of surfactant-stabilised emulsions before and after filtration: (a) water-in-oil emulsion and (b) oil-in-water emulsion (Chen et al., 2009).

environment. Korhonen, Kettunen, Ras, and Ikkala (2011) tested another CNF application as an oil absorbent by coating a CNF aerogel with a thin TiO_2 layer through a sol-gel approach. This treatment transformed the cellulose aerogel into a superhydrophobic material with a high absorption capacity for oils and nonpolar compounds. Jiang and Hsieh (2014) achieved even higher sorption capacities of model organic solvents (from 139 to 345 g g^{-1}) using CNF aerogels after modification by vapor deposition of triethoxyl(octyl)silane. The addition of the hydrophobic silanes to the CNF structure made the material oleophilic and hydrophobic, preventing oil from spreading or becoming trapped below the water surface.

9. Membranes of pervaporation separation

Pervaporation is a process for separating a liquid mixture by partial evaporation through a nonporous or porous membrane. Nanocellulose membranes have shown great potential for pervaporation separation, which is a membrane-based process used to separate liquid mixtures by preferentially permeating one component through a membrane while retaining the other components. Typically, the upstream side of the membrane is under ambient pressure and the downstream side is under vacuum to allow evaporation of the selected component after it passes through. The driving force for separation is the partial pressure difference between the components on the two sides, rather than the difference in the vibrations of the components in the feed. Nanocellulose membranes demonstrate resistance to swelling in certain solvents and can maintain their structural integrity under harsh operating conditions. This resistance to swelling and compatibility with various solvents make nanocellulose membranes suitable for separating a wide range of liquid mixtures. Fig. 10 shows the percolation and evaporation processes of nonporous membranes used to separate the liquid mixture (solvent recovery and purification) (Vane, 2020).

Hydrophilic nanocellulose membranes can be used to dehydrate alcohols containing small amounts of water, while hydrophobic membranes are used to remove/recover small amounts of organics from aqueous solutions. The principle is based on the affinity of the feed for the membrane material, where only molecules with high affinity can diffuse through the membrane (Jyoti, Keshav, & Anandkumar, 2015). Nanocellulose as a novel nanofiller has the potential to significantly improve the mechanical, electrical and thermal properties of the membrane and can solve the problem of selective pervaporation. Due to its remarkable reinforcing properties, CNC has attracted the attention of researchers. Blended membranes containing about 80 % BC exhibit a homogeneous structure (average pore size of 10.60 A) and show improved adsorption capacity (>50 %) and water vapor permeability (0.51 g cm-2 day) (Joy, George, Chirayil, & Wilson, 2020; Phisalaphong,

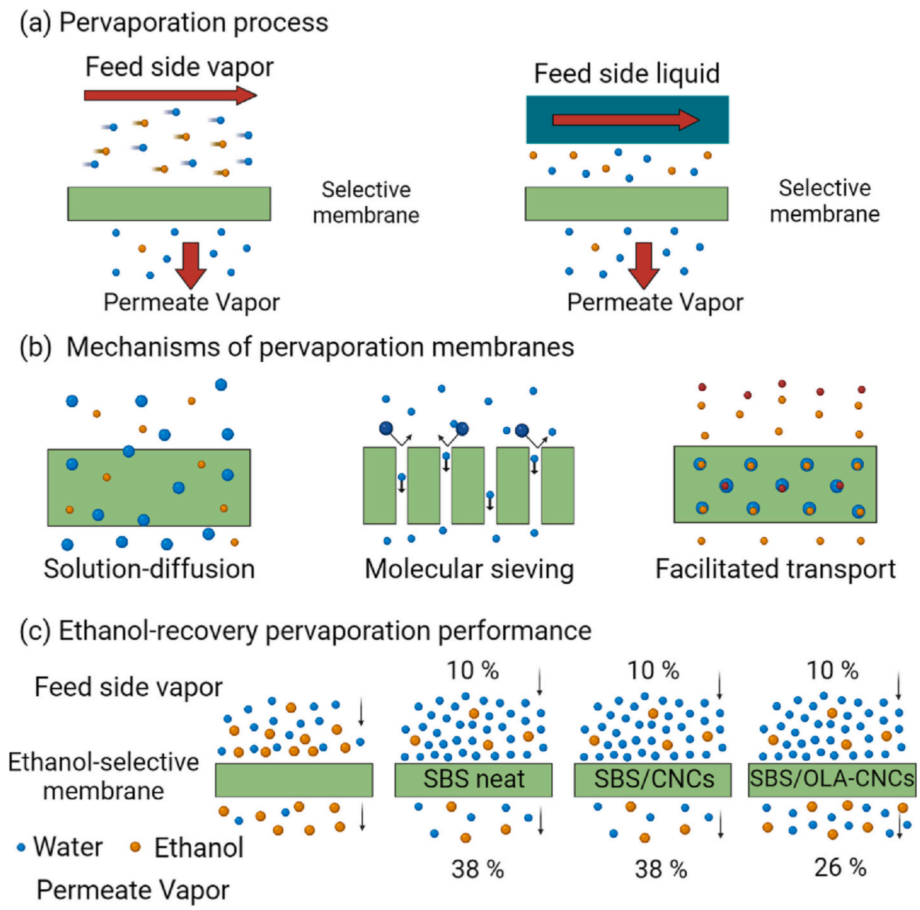

Fig. 10. Brief description of the (a) vapor permeation and (b) pervaporation processes (Vane, 2020).

Suwanmajo, & Tammarate, 2008). However, nanocellulose membranes have shown promise in mitigating fouling due to their unique surface properties, such as hydrophilicity and charge characteristics. These properties reduce the adhesion of foulants and facilitate easy cleaning, resulting in improved operational stability.

10. Water desalination membrane

Desalination is a process for obtaining highly purified water from water sources with high salinity, such as seawater, brackish water, and wastewater. Desalination is generally used to process seawater into mineral-free water suitable for human consumption (El-Dessouky & Ettouney, 2002). Membrane-based desalination is increasingly becoming a public good to produce freshwater from saline water. Physical water treatment, which enables the removal of very small impurities down to ionic size (<5 nm), can be performed using desalination membrane technologies such as RO and NF (Lee, Elam, & Darling, 2016). After the feed water contacts the membrane, the water is selectively allowed to pass while salts are retained by the membrane. RO separates salts and small molecules from low molecular weight solutes (typically <10 MWCO) at relatively high pressure (Khan et al., 2018). In a typical RO system, a pump pushes the feed water above the osmotic pressure. This allows the fresh water to pass through the membrane while all suspended solids, colloids, and soluble solids are retained. Osmotic pressure is typically between 200 and 400 psi for brackish water and between 800 and 1000 psi for seawater. The quality of the feed water (i.e., temperature, salinity, and turbidity) plays a key role in the selection of RO modules (Khan et al., 2018).

Currently, polymeric membranes dominate the water and wastewater treatment market; however, membranes made of inorganic materials (ceramic and carbon) and inorganic-organic hybrid materials can also be used. The surface hydrophobicity of polymeric membranes causes fouling, which leads to a significant reduction in flow rate, separation efficiency, and membrane lifetime when treating heavily polluted water sources (Tshikovhi, Mishra, & Mishra, 2020). On the other hand, the use of inorganic membranes is limited due to their fragility and high operating costs. The existing mineral membranes have many disadvantages, including high production cost and high working pressures. Therefore, the selection of the raw material or filler used for membrane production and modification is crucial to obtain the most suitable membranes for various purposes (Jaafar & Nasir, 2022). For practical applications, it is necessary to prepare more resistant membranes with high permeability and selectivity and better mechanical stability (Setiawan et al., 2017). These shortcomings can be mitigated by using NC materials. Pure cellulose, cellulose derivatives, and nanocelluloses have been tested for membrane fabrication because of their nontoxicity, high availability, biocompatibility, and renewability (Shaghaleh, Xu, & Wang, 2018; Tshikovhi, Mishra, & Mishra, 2020).

BC, a cheap and environmentally friendly carbon source, can be used as a carbon precursor for water desalination by the capacitive method with unprecedented desalination capacities. Nanocarbons prepared from BC have a 3D structure with hierarchical porosity and different density (Belaustegui et al., 2020). Nanocellulose membranes with a special structure have a high potential for desalination and can reduce the salinity of seawater. Liu et al. (2019) investigated CNFs as a nontoxic and low-cost filler to improve the performance of RO membranes for water desalination. They incorporated CNFs into the polyamide layer of RO membranes by introducing them directly into the aqueous phase of surface polymerization (Fig. 11). They tested the effects of different CNF concentrations on TFC RO membrane morphology, chlorine

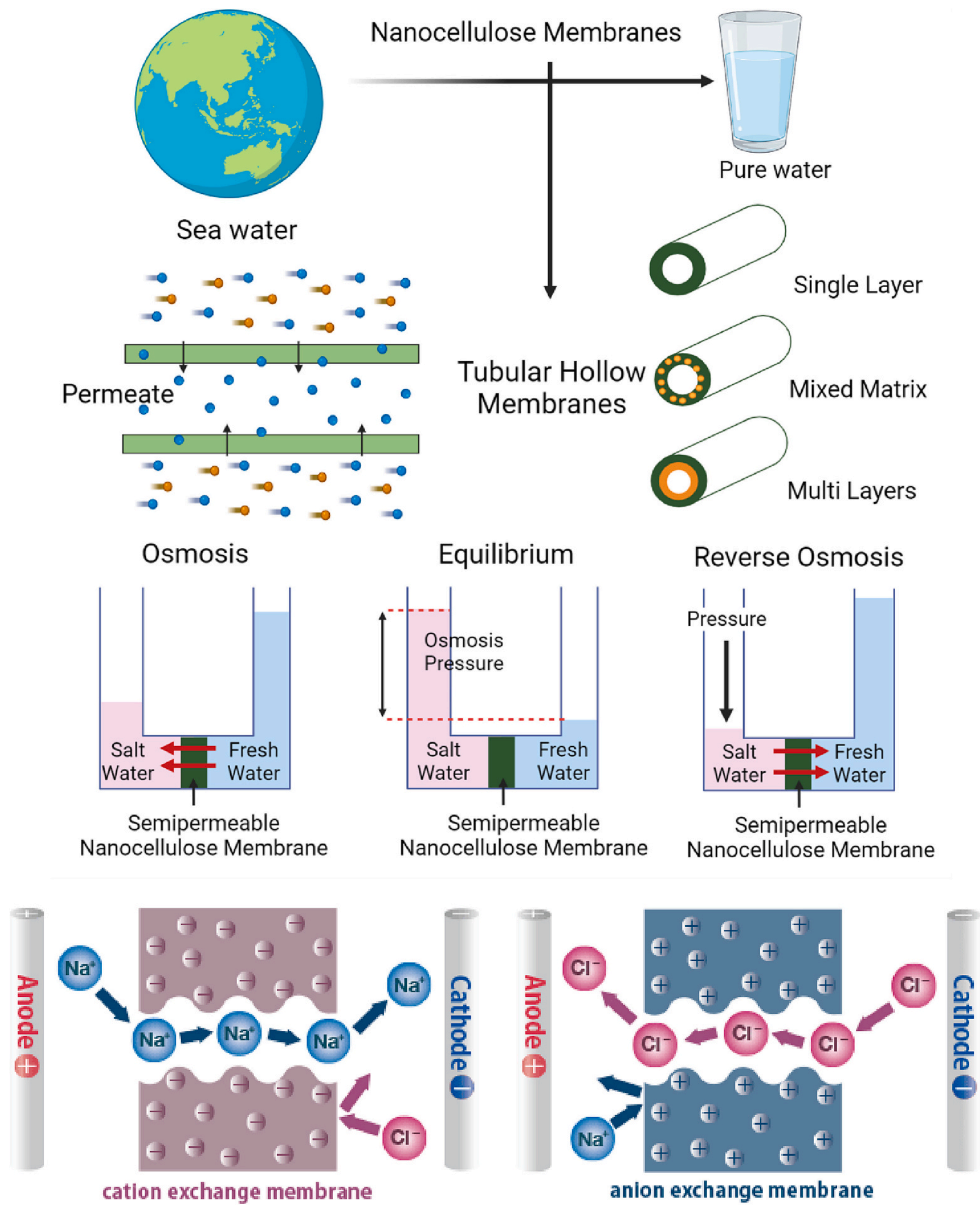

Fig. 11. Effect of CNFs on the desalination performance of thin-film nanocomposite membrane. Schematic representation of the CNF-TFC membrane fabricated by interfacial polymerization (Liu et al., 2019). **S**chematic separation process: a) cation exchange membranes, and b) anion exchange membranes (Vaselbehagh, 2017).

resistance, and desalination performance. They found that the permeability of the CNF-TFC membranes increased as a function of CNF concentration. The addition of 0.02 wt% CNFs increased the water flux of the membrane by >50 % to a value of 29.8 l m^{-2} compared to the unmodified RO membrane, and the NaCl rejection increased from 94 % to 96.2 %. These results can be attributed to the increase in membrane moisture and the thinner selective layer due to the addition of CNFs (Liu et al., 2019).

UF membranes prepared by CNF and CNC casting on PAN/PET scaffold showed high performance in oily water. The presence of CNF and silica nanoparticles on polyamide-amine-epichlorohydrin (PAE) as a composite membrane led to better UF results. In addition, CNF is a good substitute for a nanofiller and can be added to a polyamide/polysulfone RO membrane by interfacial polymerization. CNC with hydroxyl group was used as a crosslinking agent between PAN (skin layer) and polyethersulfone (support layer) in NF membranes and enhanced NaCl and Na_2SO_4 retention performance to 22.7 % and 98 % (with increasing amounts of CNC content), respectively (Tan et al., 2020a, 2020b). The chemical modification significantly improve the performance of nanocellulose as functional membrane materials for water desalination (Rana et al., 2021).

11. Electrically driven (ED) membranes

Electrically operated (ED) membranes are generally used for water/wastewater treatment by electrically operated reverse membrane processes (EDR). ED and EDR membrane processes use the difference in electrical potential to cause soluble ions to migrate through the water-impermeable membrane. When a voltage is applied to the electrodes, direct current (DC) is generated and the cations are forced to move toward the cathode. The most important property of ion exchange membranes is their ionic permselectivity, which helps classifying them into cation exchange and anion exchange membranes. The cation exchange membrane consists of negatively charged groups that reject anions and prevent them from passing through the cation exchange membrane, as shown on the left side of Fig. 11. This shows that cation exchange membranes are permeable only to cations. Anion exchange membranes work in reverse compared to the cation exchange membranes on the right side of Fig. 11. The result is a "dilute" stream with a lower salt concentration and a "concentrated" stream with a higher salt content than that of the feed water. With ED membranes, the flow direction is always the same, whereas, with EDR membranes, the flow direction must be reversed several times per hour. This is usually accomplished by reversing the polarity of the voltage applied to the electrodes, which causes the ion to change direction. The change in membrane pair results in an electrical scaling of the ions on the membrane surfaces. This periodic scavenging controls the formation and accumulation of scale and often allows the EDR membrane process to operate at higher penetration yields without the need for lower or fewer scale-inhibiting chemicals compared to ED membranes.

Electrodialysis is a membrane-based separation process that uses ion-selective membranes to selectively transport ions through the membranes under the influence of an electric field. Nanocellulose membranes can be functionalized or modified to possess ion-exchange properties, making them suitable for use as ion-selective membranes in electrodialysis. The surface chemistry and charge properties of nanocellulose can be tailored to facilitate selective transport of specific ions, allowing separation and purification of target ions from a solution. The tunable pore size and surface chemistry of nanocellulose can be optimized to selectively transport ions and exclude unwanted species. This selectivity contributes to the efficiency and effectiveness of electrodialysis processes. Nanocellulose membranes can be combined with other advanced technologies to improve the performance of electrodialysis processes. For example, combining nanocellulose membranes with nanofiltration or reverse osmosis membranes can enable multistage separations that allow more efficient and selective ion transport. Recent research showed that the deposition of various conductive materials on nanocellulose substrates by mixing, coating, or in-situ polymerization increases their electrochemical potential due to changes in the porous structure (Du, Zhang, Liu, & Deng, 2017). The presence of conductive material near nanocellulose membranes increases their flexibility and mechanical strength (especially when applied to the inner pores) and also enhances their electrochemical performance (Hsu & Zhong, 2019). Further studies are needed to optimize membrane properties, electrode configurations, and system design to realize the full potential of nanocellulose membranes in electrodialysis applications.

12. Paper filter membranes

Filter paper is a semi-permeable paper barrier that is perpendicular to the flow of liquid or air. It is used to separate tiny solid particles from liquids or gasses. Filter paper has several properties of interest, such as particle retention, volumetric flow, wet resistance, porosity, efficiency, compatibility, and capacity. Paper filtration is based on two mechanisms: surface filtration (i.e., particles remain attached to the paper surface) and volume filtration (i.e., particles remain attached to the filter paper). Filter paper is commonly used because even a small piece of filter paper absorbs a significant amount of liquid. Cellulose nanopaper membranes and filters, which are made entirely of nanoscale cellulose, are usually produced using a papermaking and filtration process (vacuum) or solvent evaporation technique. Dense nanopapers work by not absorbing contaminants larger than the pore size of the paper and by absorbing charged contaminants via electrostatic interactions. A fundamental step for the fabrication of nanocellulose -based filters and membranes is the study of thin cellulose films. The main advantages of all cellulose structures are their biodegradability and production from sustainable resources, which facilitate their disposal compared to synthetic polymers. It should be noted that random networks of nanocellulose fibrils (i.e., cellulose nanopapers made from BC or plant CNFs) can retain contaminants as large as viruses (i.e., up to 10 nm) (Wang et al., 2019). Experimental studies on this approach (Fig. 12) have shown that although these nanopaper membranes and filters have great potential for nanoparticle retention, their major drawback is their limited permeability (Wang et al., 2019). In fact, the retention performance is in the narrow UF range, while the permeance is more in the RO or at best in the NF range. Many efforts have been made to optimize the permeability of the nanopaper structure, but the results show that the retention performance decreases with increasing pore size. So far, a certain film thickness is required to ensure defect-free nanopapers and their use for nanoparticle deposition by size exclusion, which in turn limits their permeability (Mautner, n.d.).

13. Challenges in nanocellulose separation membranes

Tremendous amount of research has been carried out to optimize nanocellulose membranes for different applications such as wastewater purification, desalination, gas separation, and liquid filtration. While nanocellulose membranes hold great promise for various applications, there are still challenges that need to be addressed to fully realize their

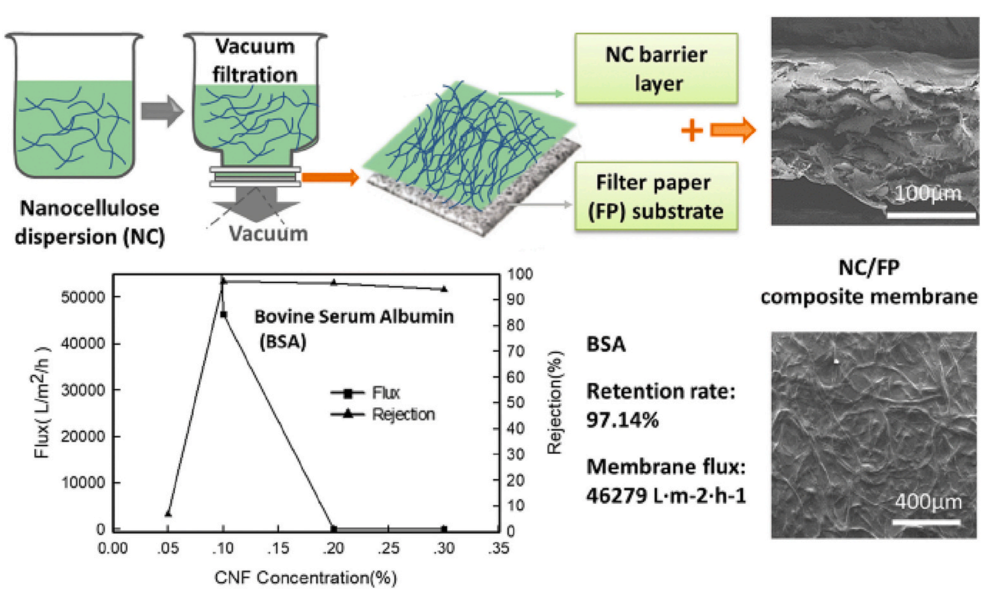

Fig. 12. Preparation of NC/filter paper (NC/FP) composite membranes for high-performance filtration (Wang et al., 2019).

potential. Some of the challenges associated with nanocellulose membranes include:

(1) Scalability and commercialization: commercialization is a main challenge as the production cost for extracting cellulose from raw materials is high. A great deal of research has been conducted to optimize the performance and production of nanocellulose. However, most of these studies are restricted to batch scale, and only a few works have been reported under the pilot scale. Thus, it is important to establish safe and cost-effective industrial methods for nanocellulose production and scale-up in order to be commercialized. Besides, maintaining the nanoscale structure of nanocellulose during handling and storage is difficult because of its tendency to reaggregate upon drying.
(2) Energy and chemical consumption: the specific energy consumption for nanocellulose production can vary depending on the manufacturing method, equipment used, and the specific application. Different processes may have different energy requirements. However, overall, nanocellulose shows promise as a sustainable and energy-efficient material with potential applications in various industries. Despite energy and time consumption of the process, to isolate the bulk amount of cellulose, the use of hazardous chemicals is harmful to the environment and human health.
(3) Mechanical strength: Nanocellulose membranes may possess excellent mechanical strength at the nanoscale, but maintaining their strength and integrity at larger scales can be challenging. Enhancing the mechanical properties of nanocellulose membranes, such as their tensile strength and flexibility, is crucial for practical applications. Nanocellulose membranes can be susceptible to water damage, especially when exposed to high humidity or immersed in liquids. Retaining their structural integrity, porosity, and mechanical properties under wet conditions is essential. Researchers are actively working on improving the water stability of nanocellulose membranes through various methods, such as surface modifications or coatings.
(4) Fouling of membranes: fouling is a common challenge in membrane-based processes, including nanocellulose membranes. This is the accumulation of undesirable substances such as particles, organic matter, or salts on the membrane surface or in the pores of the membrane. However, optimizing the membrane structure through manufacturing techniques, such as varying the nanocellulose concentration, incorporating additives, or using composite structures, can help reduce fouling. Adjusting pore size or additional surface modifications can help minimize fouling by preventing large particles from entering the membrane structure. Pretreating the feed solution or performing prefiltration steps before reaching the nanocellulose membrane can effectively reduce fouling.
(5) Reproducibility: reproducibility is a crucial aspect in the research and development of nanocellulose membranes. Cellulose sources can vary depending on factors such as geographic location and plant species. Consistent source and quality of cellulose materials is critical for reproducibility. Establishing reliable supply chains and ensuring consistent quality of cellulose feedstock are essential to maintaining reproducibility in the nanocellulose membrane manufacturing. Achieving consistent results across different batches and laboratories is essential for the widespread adoption and commercialization of nanocellulose membranes. Therefore, the development of standardized protocols for the synthesis, processing, and characterization of nanocellulose membranes is critical.
(6) Toxicity and environmental impact: Studies on the inhalation toxicology showed that nanocellulose in powder form are cytotoxic (Sharma et al., 2020). Nanocellulose membranes are considered safe for drinking water applications and the food industry, but further research is needed to obtain more evidence on this topic. Although nanocellulose membranes seem nontoxic, the studies concerning the toxicology of functionalized nanocelluloses and their nanocomposites are very few, which restricts their up-scaling implementation. Thus, from a commercial point of view, understanding the long-term toxicity of functionalized nanocelluloses and nanocelluloses nanocomposites in batch and pilot scale is necessary.

Addressing these challenges requires interdisciplinary research efforts involving materials science, chemistry, engineering, and collaboration with industry. Continued advances in nanocellulose synthesis, processing techniques, and functionalization methods will pave the way to overcome these challenges and unlock the full potential of nanocellulose membranes in various applications. By paying attention to the considerations discussed and continuously refining production processes and protocols, researchers and manufacturers can improve the reproducibility of nanocellulose membranes, leading to more reliable and consistent results in their development and application.

14. Conclusion and future prospects

Nanocellulose membranes are thin films or sheets composed of nanoscale cellulose fibers. These membranes are derived from cellulose, a natural polysaccharide found in plant cell walls, and have unique properties that make them attractive for various applications. Nanocellulose can be obtained by the mechanical or chemical decomposition of cellulose fibers, resulting in nanoscale dimensions. The two main types of nanocellulose used in membrane production are cellulose nanocrystals (CNCs) and cellulose nanofibers (CNFs). The unique properties of CNCs and CNFs, such as large surface area, high mechanical strength, biocompatibility, and renewability, make these membranes suitable for a wide range of applications. Nanocellulose membranes are used for filtration and separation processes. They can selectively filter out particles, colloids or contaminants based on their size, charge or other properties. Applications include water treatment, wastewater treatment, industrial filtration and air filtration. Nanocellulose membranes show promise in gas separation, such as CO_2 and N_2 capture, natural gas purification, and air separation. The nanoporous structure of these membranes enables selective gas permeability based on size and polarity. Due to the great potential of nanocellulose for membrane applications, biotechnology today is trying to solve the mass production of nanocellulose by bacteria (bacterial nanocellulose) and reduce environmental threats by selecting new environmentally friendly resources such as agricultural waste and sustainable technologies to replace the old-fashioned methods in the production of nanocellulose membranes. So far, various membrane production methods such as vacuum filtration, solution casting, coating techniques, melt blending, freeze drying, electrospinning, centrifugal electrospinning, and 3D printing of polymers and phase separation have been studied and their advantages and disadvantages have been described. However, the main challenges include commercial production, toxicity, reducing fouling in nanocellulose membranes, reducing the use of harsh chemicals in the manufacturing process, controlling the tendency of nanocellulose to aggregate by drying, and consistency for industrial production. Due to the versatile properties of nanocellulose membranes (used individually or in a composite), promising research should be conducted to overcome their weaknesses and expand the industrial and domestic uses of nanocellulose membranes. Future prospects for nanocellulose membranes involve addressing challenges related to large-scale production, cost-effectiveness, and commercialization. Efforts are being made to develop scalable and cost-efficient manufacturing processes to meet industrial demands. Collaboration between academia, industry, and government agencies is crucial to drive the commercialization of nanocellulose membranes.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Ahmed Barhoum reports financial support was provided by Dublin City University. Ahmed Barhoum reports a relationship with Dublin City University that includes: employment.

Data availability

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

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