

# Cellulose Nanostructure-Based Membranes: Structure, Synthesis, and Applications

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## Research Article

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# Abstract

Cellulose nanostructures and composite membranes incorporating nanoscale cellulose have gained attention due to their promising utility in various applications. Cellulose nanocomposites in the forms of aerogels, thin films, and nanopapers have been studied extensively. This review focuses on nanocomposite membranes of cellulose which find uses in desalination, sensing, electrical, and biomedical applications. The presence of abundant hydrogen bonds and easily functionalizable surfaces makes cellulose an appropriate material for the fabrication of membranes that can be leveraged for their adsorptive capabilities applicable to water treatment. Membranes of nanocellulose, their modifications by various reinforcements, augmentation of thermal stabilities, selectivity as sensors etc., are discussed in detail here along with pertinent physicochemical aspects. Regenerated cellulose which has different characteristics compared to natural cellulose, is also being discussed.

## 1. Introduction

Cellulose is an inexhaustible form of polymer suitable for potential applications in important areas such as healthcare, electronics, and photovoltaics (Trache et al. 2017; Eichhorn 2018; Raghav et al. 2021). Cellulose, when broken down into the nanometer dimensions, forms cellulose nanocrystals (CNCs), a highly crystalline form of nanocellulose (NC) (Trache et al. 2017). There has been a recent surge in research for sustainable and cost-efficient technology in the fields like water decontamination. Polymeric membranes dominate the current market, and these membranes are generally composed of inorganic and inorganic-organic hybrid materials (Tan et al. 2020). Incorporating cellulose either fully or partially into membranes enables membrane reinforcement due to the extraordinary mechanical and chemical properties of nanocellulose. Other non-cellulosic biomaterials like collagen and chitosan are also being studied for their use in membrane technology (Galiano et al. 2018). There has been an increasing demand for clean and efficient energy sources and fuel cells with polymer electrolyte membranes, where incorporation of cellulose has become significant (Muhmed et al. 2020).

Membranes and the study of osmotic phenomenon dates back to the 18th century. Nollet is credited with discovering the relation between osmotic phenomenon and semipermeable membranes (Lonsdale 1982). Graham performed more systematic studies to understand the diffusion of gases (Graham 1833). Earlier studies were primarily based on membranes derived from natural sources. Traube and coworkers first synthesized an artificial semipermeable membrane by using porous porcelain, which has cupric ferrocyanide precipitated on it (Mason 1991). Pfeffer (Liu 2019), Fick (Fick 1855), Van't Hoff (van't Hoff 1888), Nernst and Planck gave substantial contributions to developing various theories which accelerated the development of membrane and membrane technology (Strathmann 2006). The advent of the twentieth century saw a sudden shift in membrane technology in terms of synthesis methods and materials being used. The successful development of a working hemodialyzer in the mid-1900s paved the way for membranes to be used extensively in biomedical applications (Kolff et al. 2009). If membranes were initially developed for laboratory uses, they were soon adapted for industrial purposes. Studies incorporating nanocellulose into membrane material started in the early 1990's (Mautner 2020).

Later, two approaches evolved for using nanocellulose in membranes; (i) using a structure wholly made out of NC and (ii) a hybrid system in which NCs are integrated to a porous substrate. These membranes were used for water treatment, biomedical, and energy storage applications.

Cellulose nanostructures and membranes from cellulose nanostructures are a rapidly growing research area. An upward trend has been observed in the publication of research articles in the past 20 years (Fig. 1).

## **2. Structure, Classification, And Characterization Of Cellulose Nanostructures.**

Cellulose is a biopolymer composed of D-glucopyranose ring units linked together by beta-1,4-glycosidic bonds (Heinze 2015). These units have a chair conformation. Glucose residues are the repeating unit of cellulose (French 2017). Each of the units have three reactive hydroxyl groups; one primary and two secondary groups (Kargarzadeh et al. 2017, Klemm et al. 2005) (Fig. 2). Cellulose contains both intramolecular and intermolecular hydrogen bonds, which influences its solubility. The cellulose chain has a reducing (a D-glucopyranose ring in equilibrium with the aldehyde group) and a non-reducing end (anomeric carbon linked by glycosidic bonds) (Heinze 2015). The degree of polymerization and polymeric chain length determines the cellulosic characteristics (Trache et al. 2020). The degree of polymerization (DP) can be up to 20,000 depending on the cellulose source (Klemm et al. 2018). There are mainly four types of cellulose crystals: Cellulose I, Cellulose II, Cellulose III, and Cellulose IV. The varying unit cell dimensions and the methods for synthesis also are different. The types, synthesis methods and unit cell dimensions are tabulated in Table 1.

Table 1  
Cellulose crystal types, synthesis methods and their unit cell dimensions

Crystal type	Synthesis method	Unit cell and dimensions	ref.
Cellulose I	Natural sources like wood, straw, cotton, plant extracts	Triclinic P1 (a = 0.672 nm, b = 0.596 nm, c = 1.040 nm, $\alpha = 118.1^\circ$ , $\beta = 114.8^\circ$ , $\gamma = 80.4^\circ$ )  Monoclinic P2 <sub>1</sub> (a = 0.778 nm, b = 0.820 nm, c = 1.038 nm, $\gamma = 96.5^\circ$ )	(Nishiyama et al. 2002, 2003)
Cellulose II	Chemical regeneration and mercerisation of natural cellulose	Monoclinic P2 <sub>1</sub> (a = 0.810 nm, b = 0.903 nm, c = 1.031 nm, $\gamma = 117.1^\circ$ )	(Langan et al. 2001; Gong et al. 2017)
Cellulose III	Liquid ammonia treatment of cellulose I and II	Monoclinic P2 <sub>1</sub> (a = 0.445 nm, b = 0.764 nm, c = 1.036 nm, $\alpha = \beta = 90^\circ$ , $\gamma = 106.96^\circ$ )	(Wada et al. 2004; Wada et al. 2009)
Cellulose IV	Heating cellulose I with glycerol	Orthogonal (a = 0.803 nm, b = 0.813 nm, c = 1.034 nm)	(Gardiner and Sarko 1985)

## 2.1 Sources of Cellulose

Cellulose is found in plants, animals, algae, fungi and bacteria. It is the major structural component in plants, primarily found in combination with lignin, hemicellulose, pectin, and other substances. Various methods can be employed for the extraction of cellulose from different sources. Pretreatment, mechanical processing, and chemical hydrolysis are identified as critical steps for extracting cellulose (Khui et al. 2021). Once isolated, they can be modified and used for various applications (Table 2).

Table 2  
Sources of cellulose with applications

Source	Species	Application	ref.
<b>Wood</b>	Teak	Removal of cationic dyes like methylene blue and malachite green	(Olivera et al. 2018)
	Acacia	-	(Taflick et al. 2017)
	Rubber	Poly(lactic acid (PLA) based composite films	(Ou et al. 2021)
	Pine	Production of polymer composites	(Ditzel et al. 2017)
<b>Cane</b>	Bamboo	Biodegradable composites	(Guimaraes et al. 2015)
	Sugarcane bagasse	Bio-nanocomposite films, tissue engineering scaffold synthesis, Poly(vinyl alcohol (PVA) based films	(El Achaby et al. 2017; Lam et al. 2017; Pavalaydon et al. 2021)
<b>Seed/ fruit</b>	Cotton	Wound dressing, composite film fabrication	(Bhowmik et al. 2017), (Jiang et al. 2021)
	Coir	Poly(vinyl alcohol (PVA) based films	(Pavalaydon et al. 2021)
	Banana peel	Bio-adsorbent for enhancing bio-sorption of dyes	(Abdelghaffar 2021)
	Oil palm	Cellulose membrane for dye waste water treatment	(Teow et al. 2020)
<b>Straw Leaf</b>	Rice	Supercapacitor, bioplastics	(Agustin et al. 2014; Gao et al. 2018; Kim et al. 2020)
	Wheat	Catalytic conversion to valuable products like Syringaldehyde and Vanillin	(Yu et al. 2021)
	Pineapple leaf	Biomedical applications	(Cherian et al. 2010)
	<i>Carex meyeriana</i> Kunth	Methylene blue adsorption	(Yang et al. 2017)
<b>Bast</b>	Flax	CNC films	(Csiszár and Nagy 2017)
	Coconut husk	Poly(vinyl alcohol based hydrogels	(Thinkohkaew et al. 2020)
	Kenaf	Drug delivery excipient for curcumin	(Zainuddin et al. 2017)
	Pistachio shells	Pickering agent	(Kasiri and Fathi 2018)

Source	Species	Application	ref.
Trunk/stem	Banana stem	Biodegradable cellulose films	(Ai et al. 2021)
	Okra	Removal and recovery of Cd and Pb ions	(I. A. Okoro and O. K. Amadi 2012)
	Cassava stem	Dye removal from water	(Obele et al. 2021)
Bacteria	<i>Komagataeibacter xylinus</i>	Wound dressings, food packaging	(Sampaio et al. 2016; dos Santos et al. 2018)
Algae	<i>Valonia ventricosa</i>	-	(Bourret et al. 1972)
	<i>Valonia macrophysa</i>	-	(Chanzy and Henrissat 1985)

Plant cellulose is elementary crystallites organized parallel to each other, joined into fibres by hydrogen bonding. The elementary fibril, which is the basic fibrillar unit of the cellulose chains, are about 100 nm in length. Microfibrils, which are elementary fibrils assembled as fibrillar bundles, have 10–30 nm widths. Fibers vary in their structure and morphology according to their source. Plant sources that are extensively studied include wood (Rashad et al. 2017; Sullivan et al. 2018; Keplinger et al. 2021), rice husk (Kalita et al. 2015; Basu et al. 2019; Shahi et al. 2021), flax (Tang et al. 2019), coconut husk (Chua et al. 2020), hemp (Padinjakkara et al. 2020), sisal (Mondragon et al. 2018) and kenaf (Oyekanmi et al. 2021). Cotton fibers are about 99% of cellulose, making them a rich source of cellulose.

Algae are good cellulosic sources. Among the wide range of algae, green algae are mostly preferred for cellulose extraction. Some examples of cellulose producing algae include *Valonia* (Zanchetta et al. 2021), *Micrasterias denticulate* (Hanley et al. 1997), *Micrasterias rotate* (Kim et al. 1996), *Cladophora* (Zhou et al. 2019), *Boerogesenia* (Kargarzadeh et al. 2017). Cellulose derived from algae are highly crystalline, especially those derived from *Valonia* or *Cladophora* (Ferraz et al. 2013).

Bacterial cellulose is a promising biopolymer source which, due to its highly crystalline nature, containing no lignin, pectin, hemicellulose, provide for a very pure material. Its diameter ranges from 20–100 nm and is a three-dimensional network of ultrafine cellulose fibrils (Manoukian et al. 2019). Some bacterial cellulose sources include *Komagataeibacter xylinus* (Ho Jin et al. 2019), one of the most prominent bacterial nanocellulose (BC) producers, belonging to the group of acetic acid bacteria (AAB) (dos Santos et al. 2018). The AAB group is an essential class as these are food-grade bacteria and are generally considered safe. Other group species include *Komagataeibacter hansenii* (Güzel and Akpınar 2020), *Komagataeibacter medellinensis* (Molina-Ramírez et al. 2020), *Komagataeibacter uvaceti* (Nascimento et al. 2021), *Komagataeibacter oboediens* (Taweecheep et al. 2019), *Komagataeibacter rhaeticus* (Semjonovs et al. 2017), *Komagataeibacter saccharivorans* (Gayathri and Srinikethan 2019) and *Komagataeibacter pomaceti* (Gorgieva and Trček 2019). The bacterial cellulose has a better water retention capacity when compared to plant cellulose, thereby allowing it to be used as a structural component in biomedical applications. Also, they are highly crystalline while retaining their moldability

and flexibility. BNC does not exhibit antimicrobial activity on its own but, when integrated with other functional materials, can inhibit microbial growth (dos Santos et al. 2018). Tunicates, a kind of marine invertebrate sea animal, also produce cellulose in large quantities. The structure and properties of cellulose so created will differ depending on the species (George and Sabapathi 2015).

## 2.2 Regenerated cellulose

Regenerated cellulose is a type of material produced by converting natural cellulose to a soluble cellulosic derivative and then restoring it, usually into a fiber (e.g., rayon) or a film (e.g., cellophane) (Niaounakis 2017). High-purity cellulose is a requisite for the production of regenerated cellulose and cellulosic derivatives (Bajpai 2018). Regenerated cellulose is distinct from natural cellulose in its molecular weight, crystallinity and polymerization degree. The tensile strength and Young's modulus of regenerated cellulose nanofiber film is less than the usual CNF film. Therefore, using the latter is better for preparing nanocomposites (Zhai et al. 2021). Low thermal stability of the regenerated cellulose membrane (RCM) to treated cellulose microfibrils (t-CMF) was observed, and was attributed to the low crystallinity of RCM (Mohamed et al. 2015). The RCM extracted from recycled newspapers is an excellent alternative to paper recycling. This was demonstrated by comparing the changes in electrical, mechanical, diffusive parameters of a highly swollen RCM modified with a dendritic molecule (thiol DAB dendrimer of generation 3) to the original regenerated cellulose (RC/4) membrane (Vázquez et al. 2015). Studies at different concentration levels for other electrolytes (NaCl, CdCl<sub>2</sub>, PbCl<sub>2</sub>) establish the use of these modified membranes for heavy metal wastewater treatment. Amine functionalized RCM exhibited good catalytic activity, indicating a favourable material for industrial catalysis (Wang et al. 2017).

## 2.3 Classification of various cellulose nanostructures

Cellulose nanomaterials are classified into two classes: i) nanostructured materials, which includes cellulose microcrystals and cellulose microfibrils, and ii) nanofibers, which consists of cellulose nanofibrils (CNFs), cellulose nanocrystals (CNCs) and bacterial nanocellulose (BNC) (Islam et al. 2014; Trache et al. 2020) (Fig. 3). The characteristics of each nanocellulose class depend on the origin of the cellulose, the isolation and preparation methods.

### 2.3.1 Cellulose nanofibrils (CNFs)

CNCs are usually rod-shaped or needle-like and have dimensions of 1–50 nm in width, and can be up to a few 100 nm in length (Fig. 4A). CNCs are highly crystalline, have a large surface area, and have excellent thermal stability and strength. There are several methods for isolating CNFs from the lignocellulosic material of plants. Lignin and hemicellulose are removed during the pre-treatment process. CNFs have a higher aspect ratio when compared to CNCs due to its large surface area and a considerable amount of hydroxyl groups on the surface, enabling modification. CNFs are much more useful in the production of thin films when compared to other cellulose derivatives as they possess high viscosity and yield stress. The three-dimensional framework enables its modification for various applications using nanoparticles like CQD's (Melvin Ng et al. 2020).

## 2.3.2 Bacterial nanocellulose (BNC)

BC has a twisted ribbon-like shape with a diameter ranging between 20 nm and 100 nm. BNC is similar to other kinds of nanocellulose, exhibiting higher purity, crystallinity and water holding capacity, resulting in enhanced thermal and mechanical strength. BNC majorly consists of pure crystalline cellulose Ia (Sampaio et al. 2016).

## 2.3.3 Cellulose nanocrystals (CNCs)

CNCs have a broad range of size distribution, differing mainly due to the different synthesis methods. They are generally rod-shaped and vary from nanometers to several micrometres, with diameters of about 5–30 nm with a low aspect ratio (Beck-Candanedo et al. 2005).

## 2.4 Morphology of cellulose-based membranes

BNC has a higher crystallinity compared to other natural cellulose sources like cotton, flax, hemp etc (Sardjono et al. 2019). The roughness, charge, hydrophilicity of the surfaces and type of membrane material determine the adhesion of microorganisms to membrane surfaces (Nguyen et al. 2012). Membranes used for water treatment applications work on the principle of adsorption. The adsorption capabilities of these membranes will depend on the nature and availability of the functional groups acting as adsorption sites. In contrast, the pore structure in the membranes will determine the accessibility to the active sites. Finger-like pore patterns and differing pore sizes were observed in the metalized nanocellulose composite- thin film composite membranes (MNC-TFC) (Cruz-Tato et al. 2017). The finger-like pore formation occurs in two steps, initiation and propagation. The first step occurs by rupturing the polymer surface and then extending towards the bottom to reach the side of the finger pore.

As most of the studies of nanocellulose membranes are directed towards water treatment applications, properties influencing water filtration is briefly discussed. Dry membrane strength, wet membrane strength, membrane pore size and membrane porosity are the four major properties to be considered while studying nanocellulose membranes (Sharma et al. 2020). Dry membrane strength deals with membrane integrity during handling. Wet membrane strength is crucial for filtration membranes, and wet strength resins and cross-linking agents can reinforce them. Generally, as pore size decreases, selectivity increases. For TEMPO-oxidised nanocellulose membranes, the average pore size has been found to be 0.47 nm, making it suitable for gas separation (Fukuzumi et al. 2011). Surface coating and templating methods can be used to control the pore sizes. For water treatment, reverse osmosis (RO) membranes with pore size in the range of 1–10 nm are used, which require high energy and high pressure (15–30 bars) (Karim et al. 2016). On the other hand, nanocellulose incorporation into RO membranes can lead to the formation of interconnected “direct water channels” formed between the fibers and the matrix of the barrier layer. This will cause the permeance to increase without compromising on the selectivity (Ma et al. 2012).

## 3. Preparation Of Nanocellulose-based Membranes

Typical methods for synthesizing composite membranes are interfacial polymerization, vacuum filtration, electrospinning, freeze-drying and phase inversion (Mbakop et al. 2021). Vacuum filtration, also called the 'nanopaper approach', is employed at the industry and laboratory levels to produce nanocellulose based membranes and nanopapers (Mautner et al. 2015). The freeze-drying method was used to fabricate nanoporous membranes to remove dyes from water (Karim et al. 2014). This process is not optimal for mechanical performance but provides a porous structure. Electrospinning is another technique that can generate continuous fibres with good water flux and particle rejection, making it apt for microfiltration applications (Gopakumar et al. 2017). Membranes synthesized by the interfacial polymerization technique can be used for reverse osmosis (RO) and nanofiltration (NF) (Li et al. 2018). The process involves a poly-condensation reaction between two monomers completely dissolved in two immiscible solvents to produce a thin layer at the interface of a polymer substrate (Mbakop et al. 2021). The phase inversion technique was used for the preparation of poly (vinylidene fluoride) (PVDF) composite membranes with cellulose nanocrystals blended in (Bai et al. 2012). They can be effectively used for low-pressure filtrations. The preparation techniques for NCs based composite membranes and some of the CNCs, their synthesis and applications are depicted in Fig. 4 and Table 3 respectively.

Table 3  
Type of CNCs, synthesis methods, and their applications

Type of CNCs	Reinforcement	Synthesis method	Applications	ref.
CNF	Carbon quantum dots (CQD)	• In situ synthesis of CD@CNF	Water treatment • Selective dye removal	(Ahn et al. 2021)
	MgO nanoparticles incorporated polycaprolactone (PCL)/gelatin	• Coaxial electrospinning	• Periodontal tissue regeneration	(Peng et al. 2021)
	Fe doped CQDs	• Rapid microwave-assisted method	• Colourimetric detection of H <sub>2</sub> O <sub>2</sub>  • and glucose	(Bandi et al. 2021)
	Polypyrrole	•	• Hemodialysis	(Ferraz et al. 2013)
CNC	Carbon quantum dot	Hydrothermal	• Sensing and removal of fluoride ions in ranges of 0-200 µM	(Li et al. 2020)
	Nitrogen doped CQDs	Hydrothermal	• Chemosensors for the detection of metal ions and bioimaging	(Lizundia et al. 2017)
	Poly(3-hydroxy butyrate-co-3-hydroxy hexanoate) (PHBH)	Silylation	• Bio-degradable polymers	(Xu et al. 2018)
	Poly (butylene adipate-coterphthalate) (PBAT)	Solvent casting	• Bio-degradable polymers	(Morelli et al. 2016)
	Polyurethane foam (RPUF)	ultrasonication (solvent-free)	Mechanical engineering	(Septevani et al. 2017)
	2-hydroxyethyl methacrylate (HEMA)	Acid-hydrolysis	• Mechanical engineering	(Tatsumi et al. 2014)
	Poly(methyl methacrylate) (PMMA) and Poly(butyl acrylate) (PBA)	Grafting	Solar cell	(Maxim et al. 2020)
	Polyvinil alcohol (PVA) loaded with curcumin	Solvent casting	• Wound dressing applications	(Agarwal et al. 2013)

Type of CNCs	Reinforcement	Synthesis method	Applications	ref.
BNC	Polydopamine (PDA)	In situ incorporation of PDA into BNC during its ( <i>Gluconacetobacter hansenii</i> ) bacterial mediated growth	Removal of heavy metal ions like Cd, Pb etc, along with organic pollutant dyes like methylene blue (MB), methyl orange (MO) and rhodamine 6G (R6G) at high concentrations of 40–60 ppm	(Gholami Derami et al. 2019)
	Mesoporous polydopamine(mPDA) and Pd nanoparticles	Embedding mPDA into BNC matrix during growth process of BNC to generate mPDA-BNC membrane followed by in situ growth of Pd nanoparticles	Efficient dye removal (MB removal upto 99% even at high concentrations of 300ppm)  • Waste water treatment at industrial levels	(Gholami Derami et al. 2020)
	Laccase from <i>Myceliophthora thermophila</i>	Immersion of BNC membranes into laccase enzyme solution	• Antibacterial properties in wound dressings	(Sampaio et al. 2016)
	Nisin	BNC membranes immersed into nisin solution	• Providing antimicrobial and antioxidant properties to food packaging material	(dos Santos et al. 2018)

## 4. Applications Of Cnc Based Membranes

The membranes of CNCs are studied extensively for their wide range of applications in industries, biomedical, and energy storage devices. Membranes derived from nanocellulose retain the properties of cellulose and mainly act as a substrate upon which functionalization can occur. The surfaces can be modified depending on the function to be performed (Sardjono et al. 2019). They are cost-effective, easy to fabricate and rarely release hazardous chemical effluents compared to other methods. A few of those applications are enlisted below.

### 4.1 Sensors

Sensors are devices used to detect analytes or stimuli that can be used for biomedical and industrial applications. Sensors have to be stable, ensure reproducibility, and be sensitive to measure over a range of concentrations. The hydroxyl groups present in cellulose make it suitable for sensing applications as it provides mechanical stability and flexibility (Ansari et al. 2021). The presence of extensive intrachain and interchain hydrogen bonds, hydroxyl groups, electrostatic interactions and tunable cellulose polarization make an array of stimuli-responsive cellulose nanomaterials (CNM) (Zhu et al. 2020).

Nanocellulose can be incorporated into sensor systems in different ways, and they can act as a template, a reducing agent, a dispersant, a reinforcing agent etc. (Dai et al. 2020). Nanocellulose-based membranes and nanopapers are 2D nanocomposites lightweight, flexible, thermally and chemically stable with microporous structure (Dai et al. 2020). These composites exhibit exceptional sensing capabilities by the introduction of carbon nanotubes (CNTs), carbon quantum dots (CQDs), organic dyes, metal nanoparticles etc. (Fan et al. 2020). Sensors based on films are rampant, while nanocellulose membrane-based sensors are still in the initial stages of development (Abbasi-Moayed et al. 2018) (Morales-Narváez et al. 2015).

## 4.1.1 Bio-sensors

Biosensors use a biological molecule as a recognition element. CNMs that are biocompatible and show low cytotoxicity are suitable for biological applications. Biomedical applications include using CNM based glucose monitoring methods, urea detection, drug delivery, tissue engineering etc.

The detection of glucose is vital for the control of diabetes mellitus. For instance, a hybrid cellulose nanocrystal/ magnetite biosensor was recently developed for the detection of glucose in sweat and saliva (Tracey et al. 2020). Regular strips have glucose oxidase (GOx) affixed, converting the glucose to gluconic acid with hydrogen peroxide as a by-product. The hybrid cellulose nanocrystal/ magnetite biosensor strip has magnetite nanoparticles that reduce  $H_2O_2$  and oxidize ABTS (2,2'-azino-bis(3-ethylbenzothiazoline-6-sulphonic acid), resulting in a colour change, making it a self-indicating reaction. These films can be placed on the skin or the tongue to detect glucose. A novel method for the detection of small molecules and cells was developed, where gold nanorods were incorporated with cellulose nanofiber (CNF) to form a versatile membrane by Surface-Enhanced Raman Scattering (SERS) (Zhang et al. 2018). The CNF matrix, due to its nanoporous morphology, helps retain the analyte for competent SERS detection. Other examples of optical biosensing techniques include fluorescent approaches, colorimetric methods, and bioluminescence (Golmohammadi et al. 2017). A self-assembling BC-AuNP (Bacterial cellulose- gold nanoparticle) nanocomposite recently developed, leads to the generation of a colorimetric mechanoplasmonic bioanalytical sensor, that facilitates optical detection by mechanical stimuli (Eskilson et al. 2020).

## 4.1.2 Gas sensors

Detection of gases like  $NO_2$ , ammonia and ethylene are essential for industries. Colorimetric and electrochemical sensors have been used for gas detection alone or combined with other techniques. Mixing amine-based polymers with micro fibrillated cellulose (MFC) creates a hybrid transport membrane based on nanocellulose. For instance, a study demonstrated  $CO_2$  capture from  $N_2$  and  $CH_4$ . A polyvinyl amine product, Lupamin 9095, was added to enhance the  $CO_2$  flux across the selective layer. The improvement in the  $CO_2$  adsorption rate is indicative of the increased affinity of the material with the acid component on amine functionalization (Ansaloni et al. 2017). Likewise, gas separation membranes derived from nanocellulose could successfully separate  $CO_2$  through a mobile carrier mechanism (Venturi

et al. 2019). The amino acid L-arginine blended with carboxymethylated nanofibrillated cellulose (CMC-NFC) enhanced the permeability and exhibited absolute selectivity. Zhang et al. (2017) developed a functionalized cellulose/ graphene oxide (GO) proton-conducting membrane to detect ethanol (Zhang et al. 2017). GO has a low permeation for gases like methanol, ethanol and methane, but when it covalently links with cellulose fibres, it becomes an ideal material for electrochemical gas sensing applications. A very recent study using nanocellulose/graphene oxide (GONC) hybrid membrane attached to SnO<sub>2</sub> nanosheets (NS) looked into selective H<sub>2</sub> detection (Fig. 5) (Jung and Jang 2021). The pore size of 0.3 nm led to the effective removal of large gas molecules like H<sub>2</sub>S, CO and selectively allowed the diffusion of H<sub>2</sub> molecules. The SnO<sub>2</sub> NS@GONC sensor showed greater selectivity for H<sub>2</sub> than the SnO<sub>2</sub> NS and SnO<sub>2</sub> NS@GO sensors (Fig. 6).

### 4.1.3 Chemical sensor

Cholesterol measurement is a routine test employed to check cardiovascular diseases. Chemical methods, enzymatic assays, colorimetric assays used are often tedious processes. Electrochemical sensing methods have gained prominence due to their selectivity, reproducibility, fast response and stability. Such a sensor was developed for cholesterol detection by combining both electrochemical sensing and molecular imprinting techniques (Anirudhan et al. 2018). Using glassy carbon electrode surface (GCE) with silyated graphene oxide-grafted chemically modified nanocellulose (Si-GO-g-CMNC), both cyclic voltammetry (CV) and differential pulse voltammetry (DPV) studies were conducted. CV showed a detection range of 5.18–25.9 micro mol/L and DPV, 0.6475–10.360 mmol/L. Cholesterol recovery within the scope of 98.0% indicated the reliability of the sensor. Some chemicals on prolonged exposure can cause serious health problems, for instance formaldehyde which is widely used in industries, is a dangerous air pollutant. A formaldehyde sensor based on nanofibrous polyethyleneimine (PEI)/ bacterial cellulose membranes coated with quartz crystal microbalance could detect formaldehyde in the range of 1-100 ppm at room temperature (Hu et al. 2011). Figure 7 portrays that the amount of PEI in the membrane was key to the sensing properties. The presence of large number of hydroxyl groups in BC and amine groups in PEI provides the fibers with a large number of strong hydrogen bonds. The interaction between PEI and BC prevents the aggregation of PEI and ensures a uniform coating on the surface of BC nanofibers, which can then interact with formaldehyde molecules. This is illustrated in Fig. 8.

## 4.2 Energy applications

Nanocellulose is a suitable material for energy storage applications due to its unique properties based on its intrinsic structure. They have a high Young's modulus of 1.38 GPa and a strength of 2–3 GPa. Due to extensive hydroxyl groups, chemical modifications on the surface and integration with other active materials are possible. Flexible energy storage devices are made with nanocellulose fibres as they are used to fabricate films/aerogel substrates. Direct methanol fuel cells (DMFC) are becoming popular as their use also extends to portable devices, other than having a compact design, high power density, reliability etc. Nafion membranes, which are polymeric electrolyte membranes, are currently in use but are disadvantageous. They have a high methanol crossover, leading to Pt catalyst poisoning, reducing the

electrical performance and affecting cell efficiency. Research later led to polyvinyl alcohol, a non-toxic and biodegradable polymer whose methanol barrier property was better than Nafion but had lower proton conductivity. A study used sulfosuccinate acid (SSA) to modify cellulose to form films (Seo et al. 2009). The presence of hydroxyl groups on cellulose makes it easy to modify. The increase in SSA content could add to the cellulose membranes' ion exchange capacity and proton conductivity. A study using nanostructured Bacterial cellulose-Poly(4-styrene sulfonic acid) (PSSA) composite membranes displayed through-plane proton conductivity higher than  $0.1 \text{ S cm}^{-1}$  at  $94^{\circ} \text{C}$  and 98% relative humidity (RH), which decreased to  $0.0042 \text{ S cm}^{-1}$  at 60% RH (Gadim et al. 2014). The composite membranes were produced by in situ free radical polymerization of sodium 4-styrenesulfonate using poly (ethylene glycol) diacrylate (PEGDA), as cross-linker. The high proton conductivity can be attributed to the presence of a large number of sulfonic acid groups in PSSA. The proton conductivity increases with increase in PEGDA content, as seen in Fig. 9. Also, very high temperatures tend to make Nafion and pure PSSA membranes soft, but the presence of BC gives these membranes ample mechanical strength.

Later, a simple impregnating nanocellulose membrane with SSA, which showed improved methanol barrier property and lower proton conductivity than Nafion117 was developed (Sriuangrunghamol and Chonkaew 2021). However, sulfonated-modified nanocellulose membranes have high prospects to emerge as eco-friendly polymer electrolyte membranes in future DMFC applications.

## 4.3 Electronics- related applications

Nanocellulose can be considered a highly flexible and ultra-thin substrate, supporting electronic components for various applications. Biodegradable membranes from CNC, CNF and BNC can be used in fuel cell applications (Bayer et al. 2016). The fabricated cellulose nanopapers exhibited proton conductivity dependent on relative humidity, method of preparation and temperature. The CNC paper membrane was more conductive than the CNF paper membrane, primarily due to the increased charge carriers and the hydrophilicity/ acidity of the sulfuric acid groups added during the synthesis process.

Recently, a study used nanocellulose membrane to fabricate renewable flexible electronics (Mao et al. 2021). They used evaporation induced transfer printing technology (Fig. 10) to create nanocellulose-based liquid metal (NC-LM) printed circuit as liquid metal ink cannot be directly applied onto the nanocellulose membrane. As shown in Fig. 11, the NC-LM circuits can be used as wrist bands or attached to the nails and can also be used as multi-layer circuits, layered on top of each other, as they are extremely thin. NC-LM based wearable electronics research is in its infancy and has tremendous potential as they are both economical and environmentally friendly. Supercapacitors are a significant application of nanocellulose-based electronic devices with other applications like electric vehicles, fuel cells, smart consumer electronic devices etc. (Hsu and Zhong 2019).

## 4.4 Desalination applications

Desalination and water treatment strategies are the need of the hour, especially in developing countries where real-life applications can be the difference between life and death. Nanocellulose membranes are

suitable for water treatment applications due to their non-toxicity, recyclability, inert nature, mechanical performance, sustainability and energy efficiency. They can be used as conventional paper filters while controlling their pore sizes. The membranes synthesized by Liu et al. (2019) showed superior dye rejection performance to Nylon 66 membranes with similar pore sizes (Liu et al. 2019). These CNF membranes fabricated with an ultrathin Graphene oxide (GO) barrier layer are easy to create. They are superior to the membranes currently used for water purification and food industries.

There have been numerous reviews on the use of membranes for water treatment (Cruz-Tato et al. 2017; Tan et al. 2020; Mautner 2020). Many carboxyl and hydroxyl groups on the membrane surface aids in water treatment applications, acting as adsorbents. TEMPO-oxidized cellulose nanofibers (TOCN) that used lysozymes (LYS), a natural protein as an adhesive, showed excellent results for the removal of heavy metal ions oil droplets and molecules (> 3 nm). The carboxyl groups of TOCN and the amine groups of LYS have an electrostatic interaction that enables them to form stable membranes. The amyloid-like oligomers from LYS helped the TOCN to stick together. Due to the presence of various functional groups like hydroxyl, carboxyl, amino and thiol groups, it was an efficient water purifier, especially in the removal of boron, a toxic pollutant (Huang et al. 2021).

One of the challenges of using membranes is the risk of fouling. Earlier, fouling was treated using chemical methods, which were not very efficient as it provided only temporary relief and led to increasing costs and wastage of chemicals. The self-cleaning and anti-fouling properties of nanocellulose-enabled thin-film nanofibrous composite (TFNC) ultrafiltration membranes were studied for their ultrafiltration of BSA protein solution and wastewater. The adhesion of biomolecules to the membrane surface was hindered by surface charge, which can be controlled by the degree of oxidation of CNF (Yang et al. 2021). A novel technique by Jiang et al. (2019) uses membranes created from bacterial nanocellulose (BNC) incorporated with reduced activated carbon oxide (RGO) during its growth and inhibit biofouling, “the Achilles heel of membrane processes”, also the RGO/BNC membranes showed bactericidal activity upon illumination with light (Flemming et al. 1997; Jiang et al. 2019). Forward osmosis (FO) is a technique that can be effective against fouling, as it creates a lower transmembrane pressure when compared to other conventional water treatment methods. A study used this principle of FO to deal with wastewater treatment (Cruz-Tato et al. 2017). Silver and platinum nanoparticles were added to these nanocellulose based composite membranes to enhance their properties of the membranes. Interestingly, fouling continued in the membranes to an extent, but an improvement in solute rejection and permeation of the membrane was observed. Self-cleaning membranes are important as membranes under constant use tend to have very low flux and are unsuitable for practical applications. UV assisted self-cleaning  $\text{TiO}_2$ /TCNC membranes by Zhan et al. (2018) is a sustainable and straightforward technique for removing oil from oil/water emulsions (> 99.5%) (Zhan et al. 2018). Tunicate cellulose nanocrystals (TCNC) due to their pore size and special wettability are apt for oil/water separations. On UV-light irradiation, the membranes showed improved underwater oil contact angles and water fluxes. Multifunctional materials take care of a range of problems as they present a range of properties. Yet, processing poisonous cations, anions, and oil from water have many challenges. The oils that are also a

pollutant can affect the adsorption of the membranes, affecting the removal of cations and anions directly. Since cellulose shows outstanding acid, alkali, and salt tolerances, they are suitable for synthesizing separation materials. Hydrogels have excellent water-retaining and absorbing abilities, leading researchers to combine both of their properties to fabricate a nanocellulose hydrogel coated titanate-bismuth oxide membrane for the treatment of cations, anions and oil contained in polluted water (Xiong et al. 2018). The cations were trapped in TNF's layers (titanate nanofibers). After the exchange with  $\text{Na}^+$  ions, the anions formed irreversible compounds, like  $\text{Bi}_4\text{O}_5\text{I}_2$ , by occupying the oxygen holes of delta- $\text{Bi}_2\text{O}_3$ , providing more stability. The removal efficiency of cations and anions was also observed due to the membranes' electrostatic adsorption and hydrogen bond interaction. A nanocomposite membrane that showed tremendous potential in adsorbing anionic and cationic organic dyes was developed by researchers (Vilela et al. 2019). When combined, zwitterionic poly (2-methacryloyloxyethyl phosphorylcholine) (PMPC) and bacterial nanocellulose give optically transparent nanocomposites that inhibit the growth of pathogenic bacteria. The dye removal capacity for a nanocomposite membrane containing 79 wt.% of PMPC was estimated to be  $4.44 \pm 0.32 \text{ mg g}^{-1}$  for methylene blue (cationic dye) and  $4.56 \pm 0.43 \text{ mg g}^{-1}$  for methyl orange (anionic dye) (Fig. 12).

The active area of water decontamination has a new entrant in bacterial nanocellulose aerogel membranes (Leitch et al. 2016), which was first studied for membrane distillation (MD). Ferreira-Neto et al. (2020) created aerogel membranes of bacterial nanocellulose and molybdenum disulphide ( $\text{MoS}_2$ ) nanosheets, which made it quite efficient in the removal of dyes and heavy metals (Ferreira-Neto et al. 2020). It is bifunctional, acting as both a photocatalyst and an adsorbent, unlike earlier membrane technologies that look into photocatalytic properties for in-flow water purification. The FT-IR and XRD analysis confirmed that the use of  $\text{MoS}_2$  did not compromise the structural integrity of the BC membrane up to six photocatalytic cycles. Even though a surface passivation layer of  $\text{MoO}_3$  was observed, it did not affect the overall performance of the membrane. The EDS and mass analysis showed only about  $< 0.3\%$  of total Mo leached into the solution, indicating photostability of the membrane. One of the primary challenges of MD is sustainability as they require high input of energy, while photo-thermal membrane distillation (PMD) is efficient in terms of energy. A bilayer membrane made of environmentally sustainable materials, polydopamine (PDA) and bacterial nanocellulose (BNC) was developed for PMD, which exhibited a permeate flux of  $1.0 \text{ kg m}^{-2} \text{ h}^{-1}$  under one sun irradiation (Wu et al. 2021). The membrane, fluoro-silanized using (tridecafluoro-1,1,2,2-tetrahydrooctyl)-trichlorosilane (FTCS), gives high salt rejection ( $> 99.9\%$ ) and only permits vapor transport. High porosity ( $\sim 93\%$ ) and excellent optical activity ( $\sim 98\%$ ) enhance the PMD performance. These solar-driven membranes possess bactericidal properties, minimizing bio-film formation and microbial aggregation, thereby increasing the longevity of the membrane. A state-of-the-art solar-driven sweater desalination technique was developed, which combined the two-dimensional membrane and three-dimensional foam into a single product (Zhang et al. 2020). The membrane's mechanical strength combined with the foam's high porosity makes it different from conventional nanocellulose membranes. Functionalities are introduced to the nanocellulose foam membrane (AGM) by adding carbon nanotubes (CNT). The water evaporation rate of AGM-CNT ( $1.67 \text{ kg}$

$\text{m}^{-2} \text{h}^{-1}$ ) was higher than the normal CNF-CNT ( $1.1 \text{ kg m}^{-2} \text{ h}^{-1}$ ) membranes, revealing the combined effect of the porous and hydrophilic nature of the AGM-CNT's.

## 4.5 Applications in dermatology and cosmetics

Research in dermatology and cosmetics is on the rise, especially the search for natural ingredients which are much more compatible with the skin. BC has been extensively commercialized for dermal requirements due to its high-water retention capacity and the ability to hydrate the skin (Almeida et al. 2014). A group of researchers could successfully study the effect of dry BC: CMC (Bacterial cellulose and Carboxymethyl Cellulose) incorporated into generic cosmetic creams, replacing the traditionally used surfactants (Martins et al. 2021). Both viscosity and texture of the creams could be replicated with the replacement of 5.5% surfactants with only 0.75% BC: CMC. More ingenious methods are being created to incorporate BC and other cellulosic derivatives into uses other than topical applications. More recently, researchers developed an innovative patch for dermo-cosmetic applications based on two biopolymers, hyaluronic acid (HA) and BC (Fonseca et al. 2021). The microneedle (MN) application method is durable to the human skin revealing its cutaneous compatibility. The effectiveness of the system was demonstrated using Rutin, a natural antioxidant. The peculiar porous structure, water retention ability, moldability and good mechanical properties make BC a good biopolymer for cosmetic purposes. The regenerative properties of HA and the capacity of BC to release bioactive molecules makes it suitable for skin applications. Transdermal drug delivery is another application of nanocellulose membranes. This method is now being preferred over the traditional ones as there is hardly any interaction with a systematic circulation.

A recent study demonstrated the transdermal delivery of crocin, an active agent of saffron (*Crocus sativus*), loaded into BNC membranes (Abba et al. 2019). A gradual release of crocin over 7 hours and cumulative release of  $215 \mu\text{g}/\text{cm}^2$  was observed, which can be associated with the unique morphology of BNC membranes having an entangled nanoscale structure and 3D network. Further research in this area can help include other water-soluble active compounds in transdermal delivery applications.

## 5. Conclusions And Outlook

This review has discussed nanocellulose derived membranes, their synthesis, and their applications. These membranes, which are basically made of nanocellulose, embody all the properties of nanocellulose and, when combined with other functional materials, show characteristic properties that make them a promising class of materials to be further explored. As researchers focus more on greener and sustainable alternatives, nanocellulose membranes are envisaged as promising materials in biomedical research and desalination applications. One of the significant challenges of using nanocellulose based membranes in desalination techniques is the problem of fouling. Membranes also have to undergo pretreatment before being used in practical applications. They are rarely used without modifications as structural and compositional augmentations are essential to enhance the properties of the membranes. Their reusability and reliability in the long run are yet to be studied. However, many

advancements are still in their initial stages but the membranes currently produced are sufficiently stable both chemically and thermally. One of the advantages of membrane technology based on cellulosic materials is that it is incredibly energy efficient. They have a meagre environmental impact as no hazardous chemicals are involved in most of the cases. Cellulose is environmentally friendly, and an inexhaustible source which has tremendous potential to develop into an irreplaceable material of the future.

## Declarations

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### competing interest

There are no conflicts to declare.

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## Figures

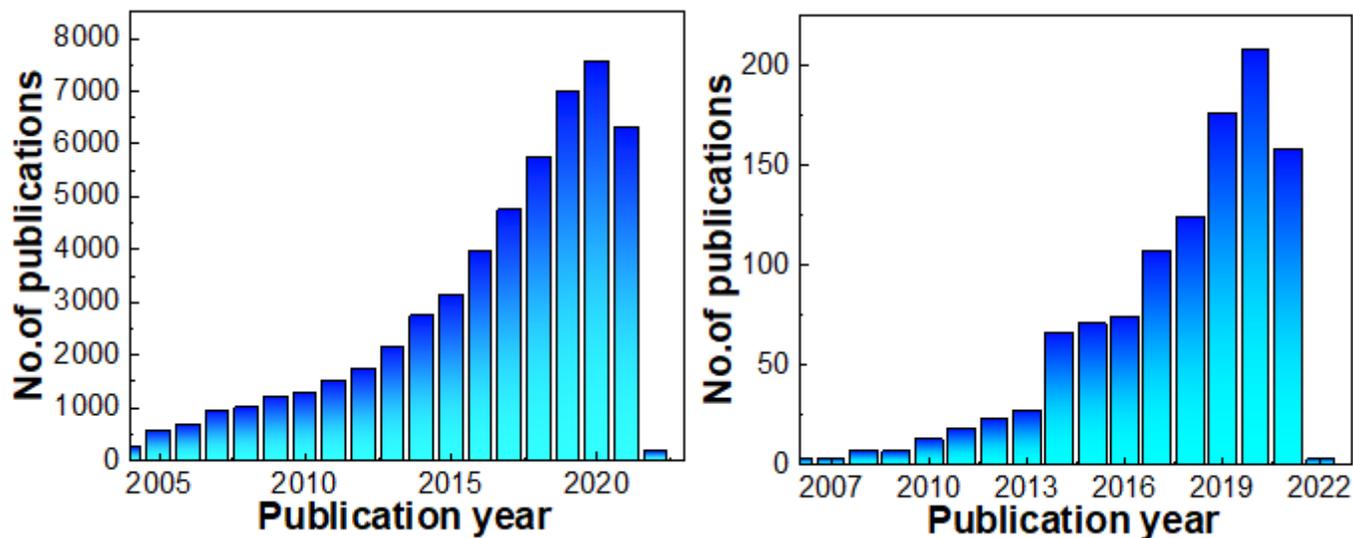


Figure 1

Statistics depicting the surge in active research on CNCs and their membranes. The literature search focused on documents containing “cellulose nanostructures” and “membranes of cellulose nanostructures” in their title, abstract, and keywords. Data courtesy: Web of Science (acquired in November 2021).

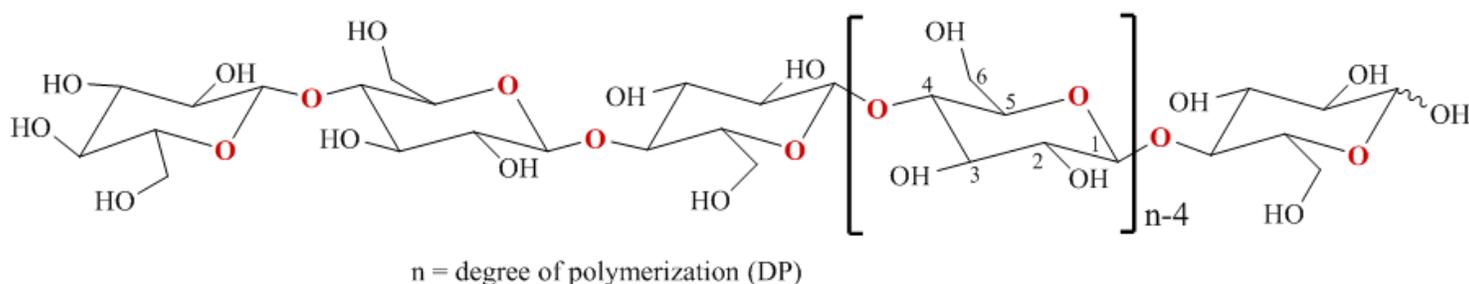
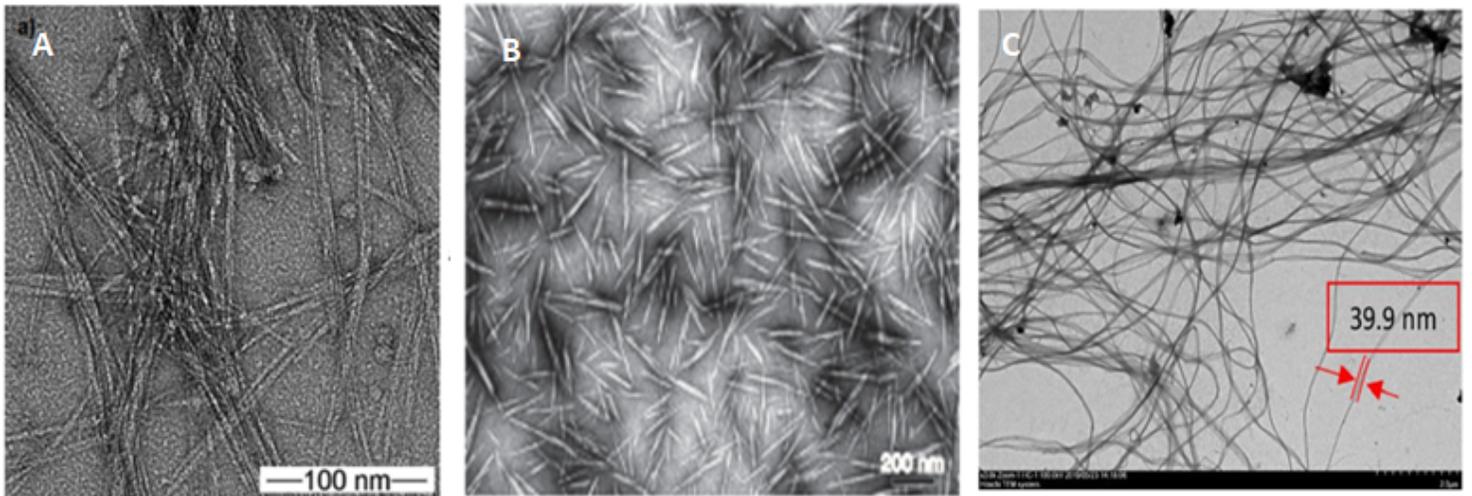


Figure 2

Molecular structure of cellulose.



**Figure 3**

A) CNF from banana peel. Reproduced with permission(Tibolla et al. 2017). Copyright 2017, Elsevier B) CNC from ramie fibre. Reproduced with permission(Tayeb et al. 2018). Copyright 2021, The Royal Society of Chemistry. C) BNC from banana peel. Reproduced with permission(Sijabat et al. 2020). Copyright 2020, IOP Publishing.

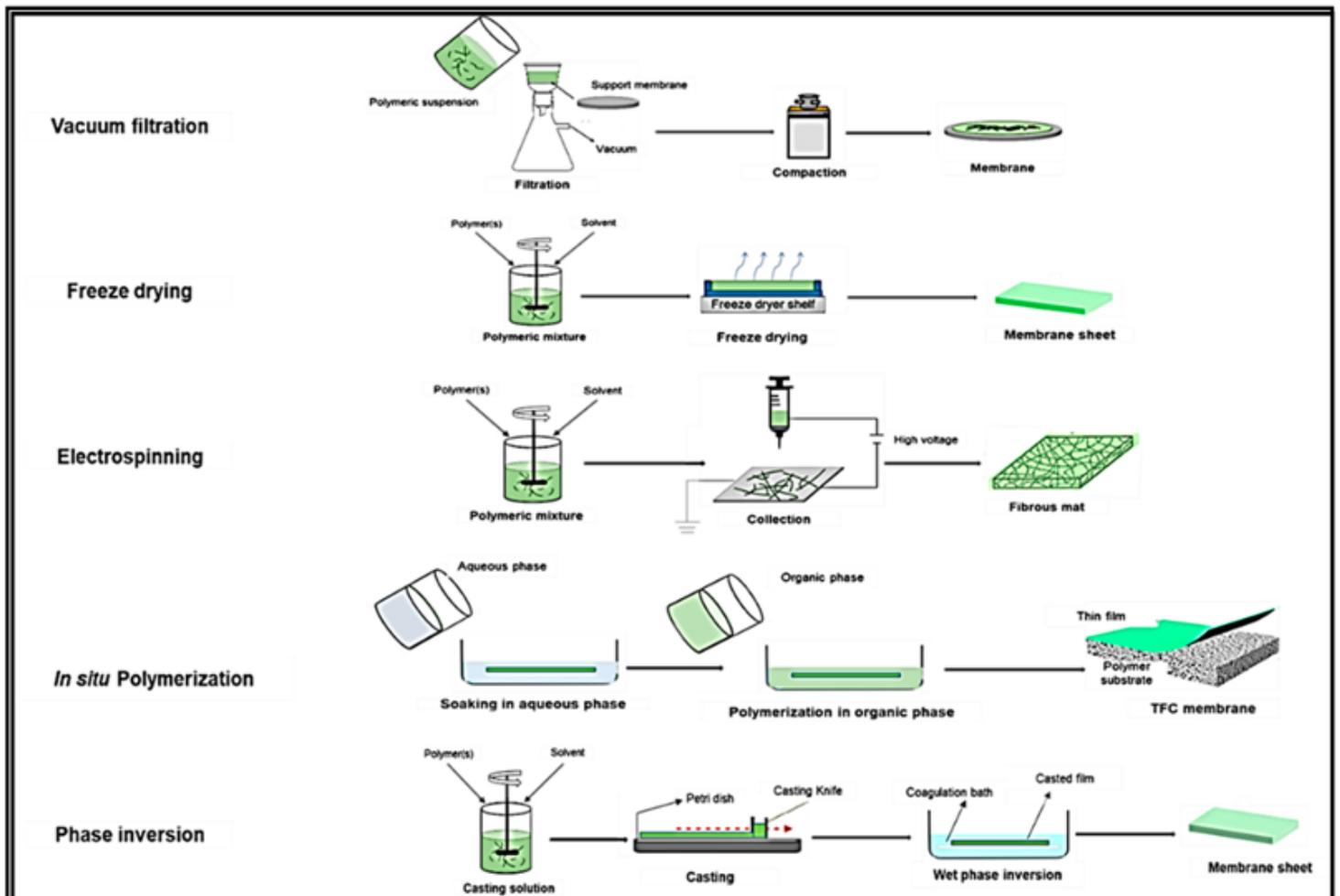


Figure 4

Schematic illustration of commonly utilized preparation techniques for NCs based composite membranes. Reproduced with permission (Mbakop et al. 2021). Copyright 2021, MDPI.

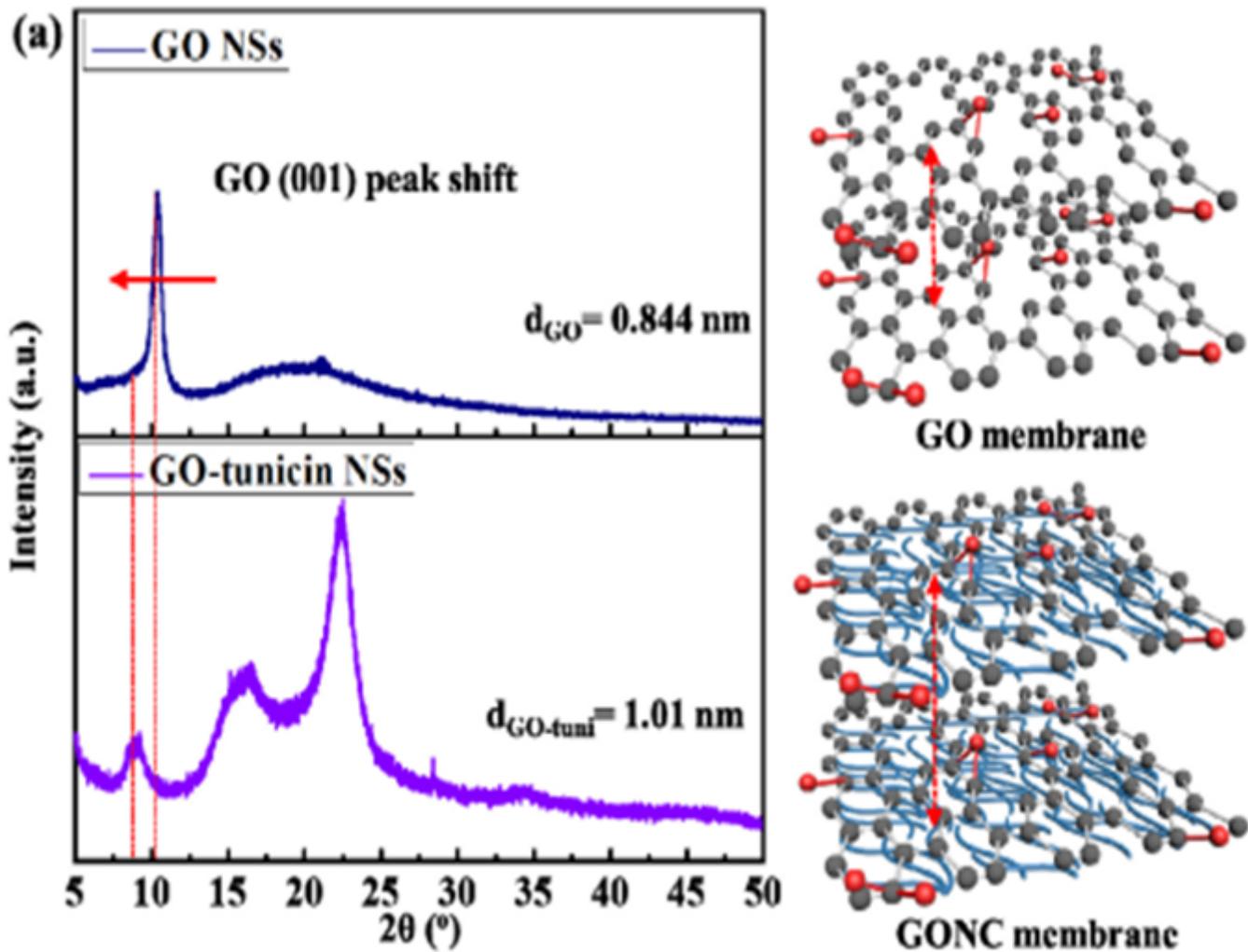


Figure 5

XRD patterns of GO and GONC with corresponding d-space values, and schematic images of GO and GONC. Reproduced with permission (Jung and Jang 2021). Copyright 2021, Korean Sensors Society.

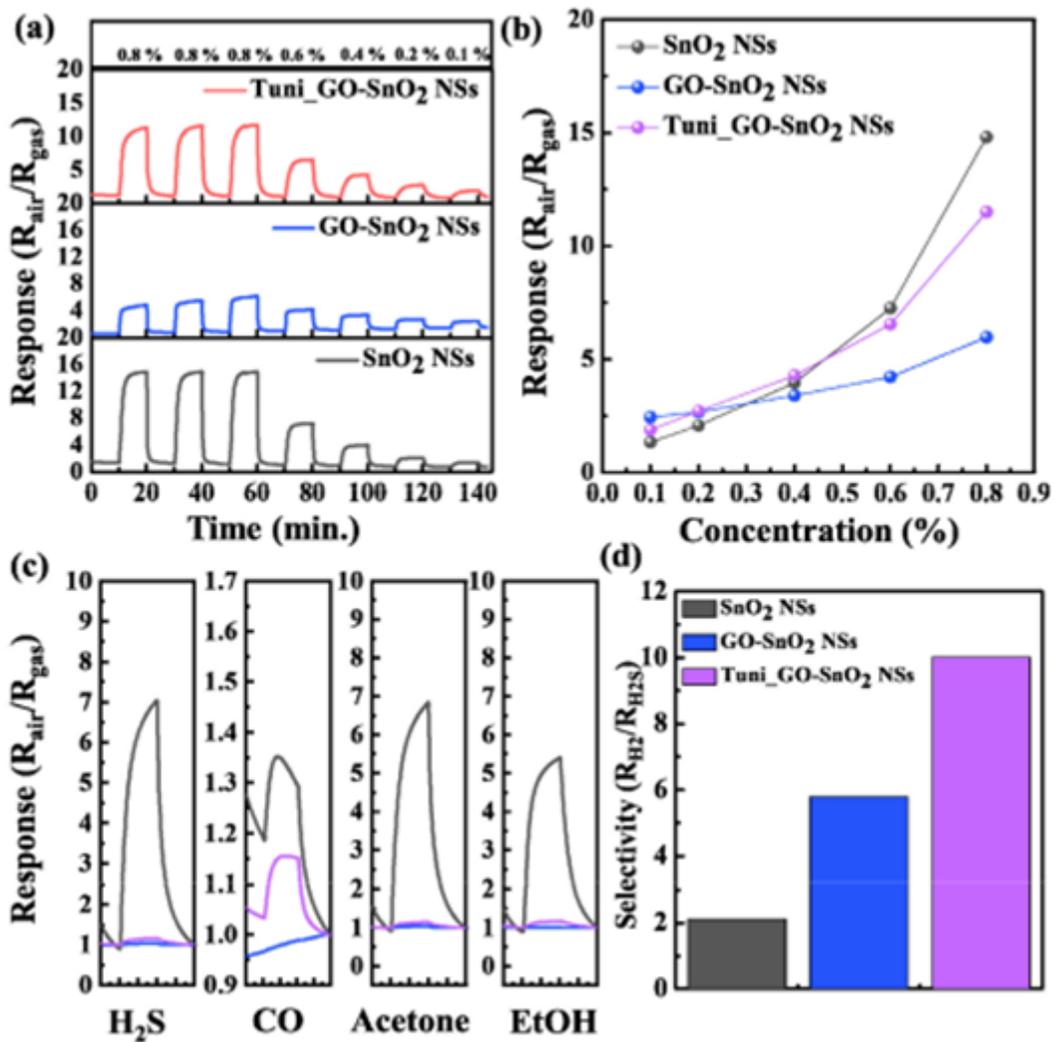
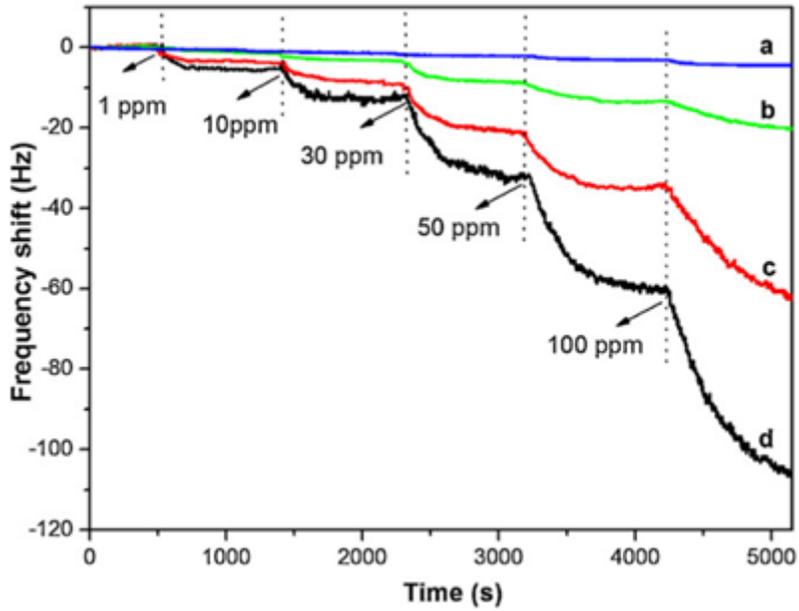


Figure 6

a) Hydrogen gas sensing properties of SnO<sub>2</sub> NS-, SnO<sub>2</sub> NSs@GO-, and SnO<sub>2</sub> NSs@GONC-based sensors, b) graph of hydrogen gas response values with respect to hydrogen gas concentration, for each sensor, c) sensing response kinetics of each sensor to H<sub>2</sub>S, CO, acetone and ethanol, d) sensing selectivity polts of SnO<sub>2</sub> NS-, SnO<sub>2</sub> NSs@GO-, and SnO<sub>2</sub> NSs@GONC-based sensors. Reproduced with permission (Jung and Jang 2021). Copyright 2021, Korean Sensors Society.



**Figure 7**

Response of sensors coated with (a) BC membrane, (b) PEI membrane, (c) PEI/BC membranes with a weight ratio of 0.67/1 and (d) PEI/BC membranes with a weight ratio of 1.34/1. Reproduced with permission (Hu et al. 2011). Copyright 2011, Elsevier.

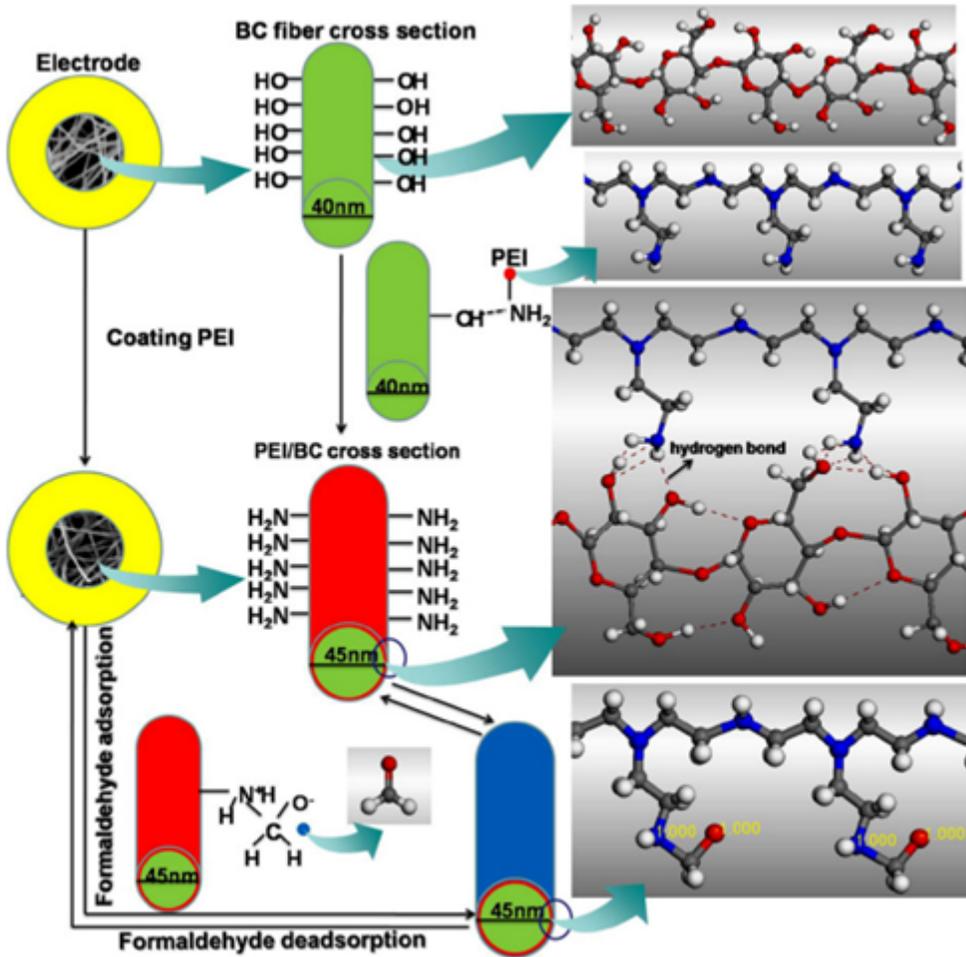
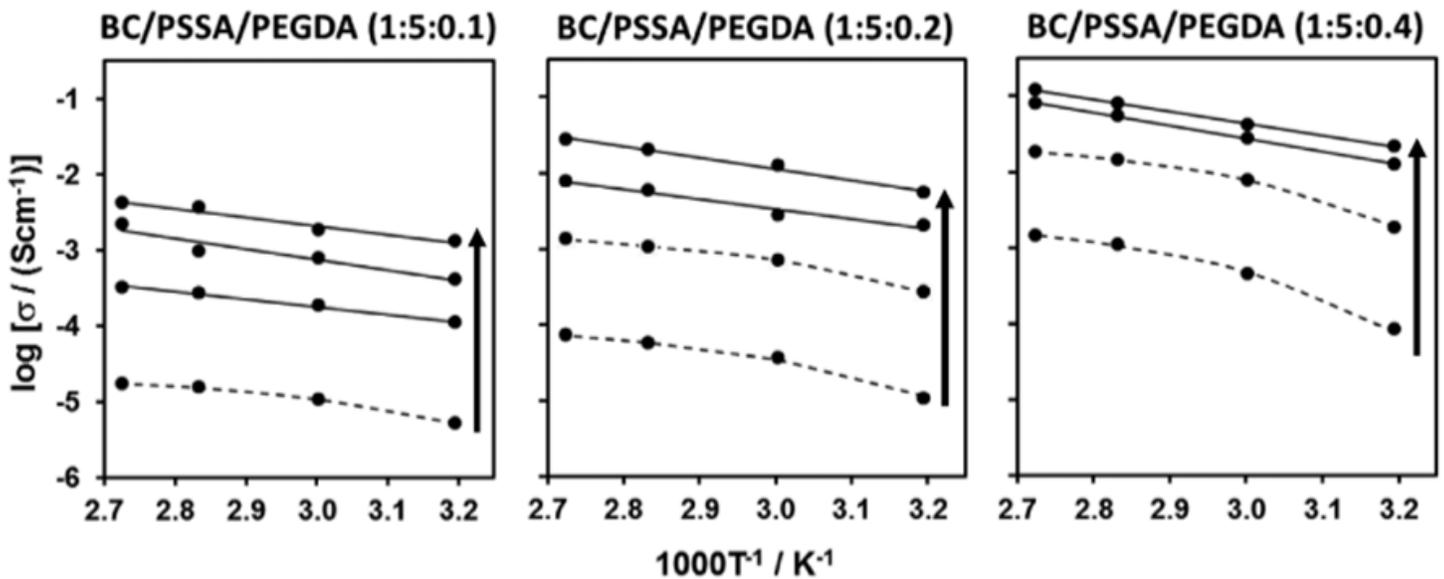


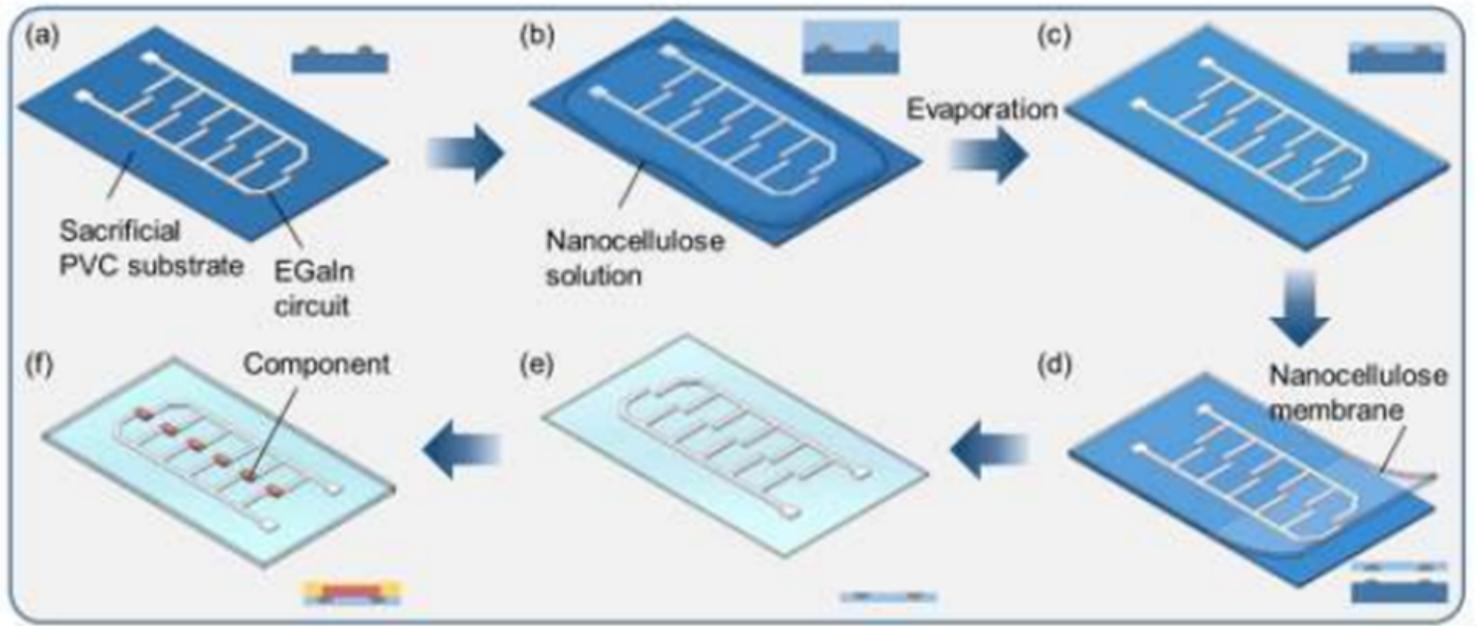
Figure 8

Schematic diagram of the interaction of BC and PEI, and formaldehyde and PEI. Reproduced with permission (Hu et al. 2011). Copyright 2011, Elsevier.



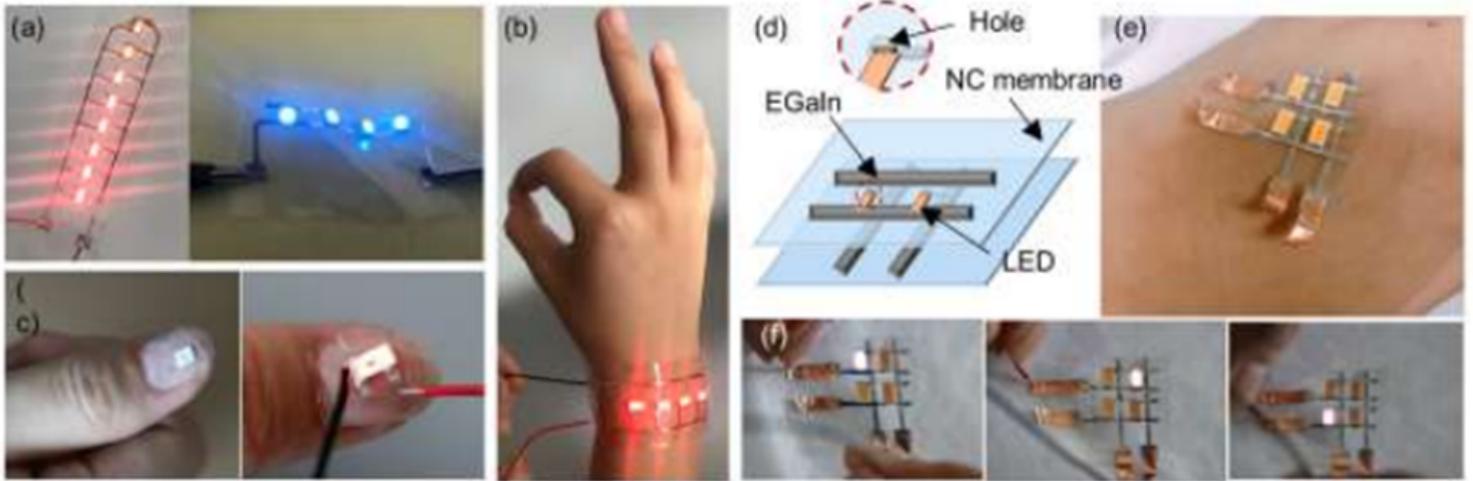
**Figure 9**

Arrhenius plots of the total conductivity of nanocomposite membranes with variable content of cross-linker measured at RH of 30%, 60%, 80% and 98% (increasing in the direction of the arrows). The straight lines are linear fits to the Arrhenius model, whereas the dashed curves are fits to the Vogel-Tamman-Fulcher (VTF) equation. Reproduced with permission (Gadim et al. 2014). Copyright 2014, American Chemical Society.



**Figure 10**

Schematic diagram of the process of the fabrication of the NC-LM circuit using the evaporation-induced transfer printing technology: a) Printing liquid metal circuit on a PVC substrate b) Pouring the nanocellulose solution onto the PVC substrate c) Evaporation and film-formation d) Peeling the NC-LM circuit off the sacrificial substrate e) NC-LM circuit with EGInk embedded in the nanocellulose membrane f) Mounting the components. Reproduced with permission (Mao et al. 2021). Copyright 2021, Elsevier.



**Figure 11**

Samples of the NC-LM circuit and its applications in the double-layer circuit fabrication: a) Flexible NC-LM circuits connected with LEDs b) NC-LM circuit rolled around the wrist as a wristband c) a tiny NC-LM circuit stuck to the fingernail compliantly d) schematic diagram of the double layer NC-LM circuit e) practical pictures of a 2\*2 LED array circuit f) performance of the LED array circuit. Reproduced with permission (Mao et al. 2021). Copyright 2021, Elsevier.

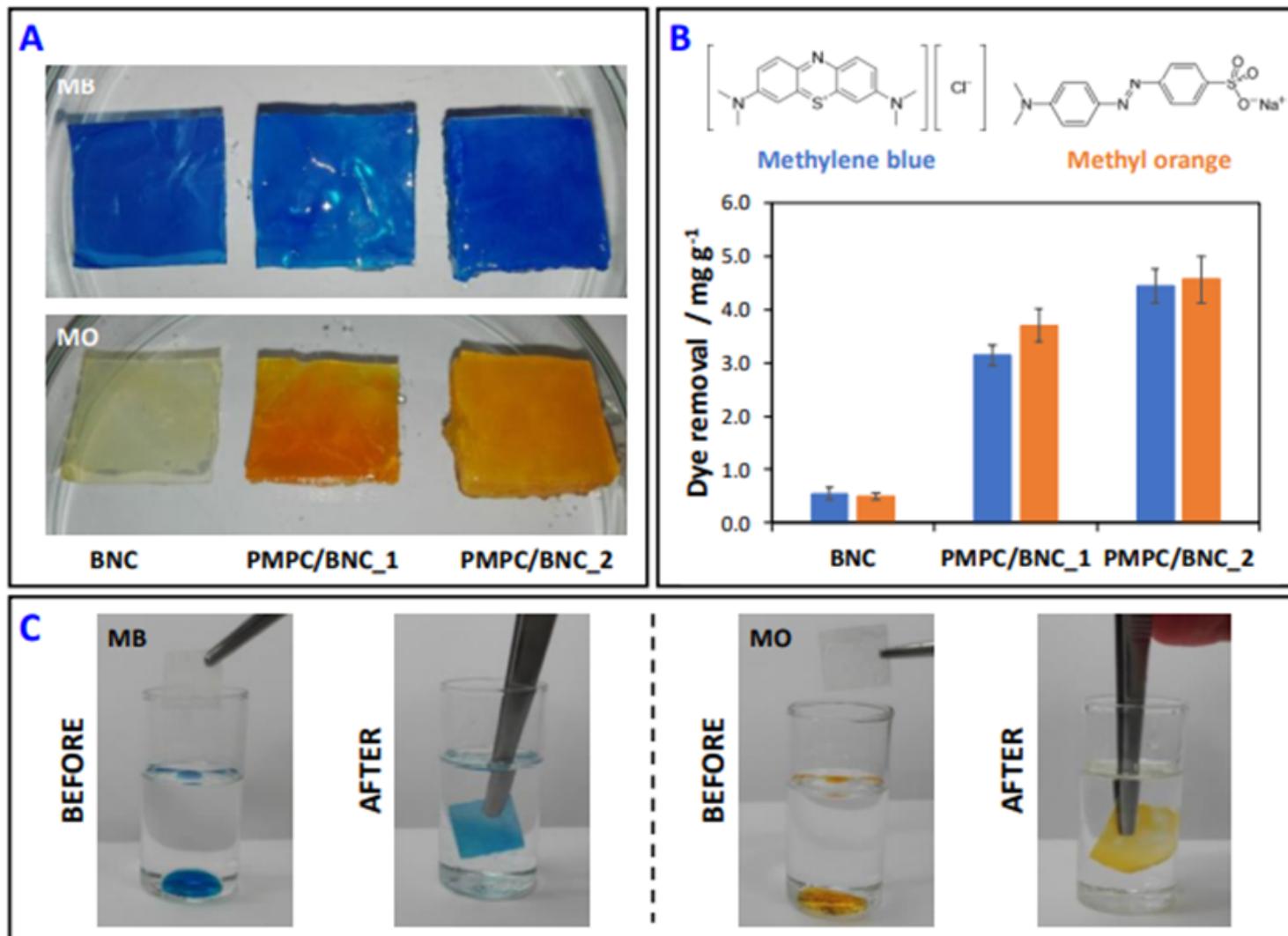


Figure 12

A, B) Photographs depicting the dye removal capacity of BNC, PMPC/BNC-1 and PMPC/BNC-2 after 12 hr immersion in dye aqueous solution and C) photograph of MB and MO aqueous solution removal from paraffin wax oil by PMPC/BNC-2. Reproduced with permission (Vilela et al. 2019). Copyright issued by creative commons license, MDPI publisher, 2019.