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Polymeric membranes as sustainable material for water desalination: a review

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ABSTRACT

Affordable potable water is the most demanded issue of whole humanity. As almost 33% of the earth is covered with seawater, which can be used with the help of various desalination techniques in sustainable commercial processes. In many commercial processes affordable and economic membranes are used which contain different types of blends that are used for desalination. The commercial or traditional membranes are synthesized and fabricated with fillers like cellulose, thin-film composite, chitosan (CS) and chitin, along with mixed oxides for filtering the sea water. To overcome the limitations of the traditional methods, pure membranes are modified precisely, which also improves their performance. Further, many researchers confirmed the mixing of 2 or 3 types of membranes to prepare a composite polymer membrane which has better performance due to higher adsorption capacity (AC). The mixed matrix membranes and metal oxide membranes generally have tunable physical properties, which influences the AC. The enhanced AC is due to high porosity, greater surface area, high permeability, high hydrophobicity, high salt rejection, and high stability (mechanical, chemical, and thermal). This chapter lists the various types of polymeric composite membranes and their performances for the desalination process.

Keywords: Desalination; Polymeric composite membranes; Mixed matrix membranes; Thin-film composite membranes; Membrane separation technology; Cellulose-based membranes; Adsorption capacity

1. Introduction

The scarcity of freshwater is rising yearly, due to population and industrial developments, which also leads to rise in global temperature [1]. Desalination is a well-known process which has potential to provide adequate freshwater for both industrial and residential purposes [2,3]. For residential usage, after desalination the water is further treated using adsorption method, chemical oxidation method, biological treatment and membrane processes [4]. The adsorption process is used to wipe-off the primarily pollutants like oil, organic matter, and grease from the seawater. The challenges for selection of appropriate adsorbents are costs, regeneration, disposal, and environmental apprehensions. The chemical oxidation process is used to disintegrate organic molecules by step-wise radical reactions. These radical reactions are carried forward by using chlorine, hydrogen peroxide, ozone, permanganate and iodine. These reactions are accelerated with UV light (photocatalytic oxidation) or ultrasound (sonochemical oxidation). The chemical oxidation treatment has its own limitations which severely affects the treatment process by lowering the radical development as well as the rate of oxidation. The oxidation reaction rate is affected by the amount of radical scavengers present, high salt levels, high chloride levels, and bicarbonate levels

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in seawaters. In case of low levels of pollutants, oxidation process becomes quite energy demanding, for example, the removal of pharmaceutical chemicals and micro-plastics. The biological methods are also actively used for the treatment of residential water, however, treating industrial water is not possible as microorganisms will not endure punitive conditions [5]. To overcome the above-mentioned limitations, the membrane technology is used for its promising features like improved selectivity, stability (thermal, chemical, and mechanical), high separation efficiency, low cost, increased life span, and low maintenance [6,7]. The added advantage of membrane technology is that it does not require chemicals and harmful by-products while treatment. For commercial purposes water desalination process is followed by thermal distillation method, multi-stage flash method, multi-effect distillation method, mechanical and thermal vapour compression methods. The desalination process is upgraded by utilizing membranes in filtration techniques, that is, ultrafiltration (UF), microfiltration (MF), reverse osmosis (RO), and nanofiltration (NF) which are adopted for large-scale industrial processes [8,9]. The commercial membrane filters have occupied big markets and are also listed in stock market dropdown list [10]. UF, RO, and MF are the most practiced processes, as compared to other pre-treatments like sedimentation and coagulation. In MF techniques, various reactors are prepared like osmotic membrane bioreactors (OMBRs) and membrane bioreactors (MBRs) for desalination and waste-water treatment whereas in traditional methods aerated lagoons, activated sludge processes (ASP) and trickling filters are used [11,12]. The active functionality component of the membrane is important for any filtration process. The membranes are made of inorganic (ceramic) materials, organic (polymeric) and biological materials depending on its applications [13]. Compared to various additives used in the membranes, the polymeric materials are most economical [14]. The polymeric membranes are widely used for seawater treatment due to its typical characteristics such as high stability (of better thermal, chemical, and mechanical) and corrosion resistance [15]. The origin of polymers is divided into two: natural (received from sources like animals and plants) and non-synthesized polymers (derived from petroleum products and synthesized in lab and/or bulk scale) [16]. The studies have come up with novel polymer membranes with high efficiency which are made with natural sources like chitin, cellulose, and starch [17]. Due to their potential high stability and reusability, inorganic membranes are well employed in membrane blending.

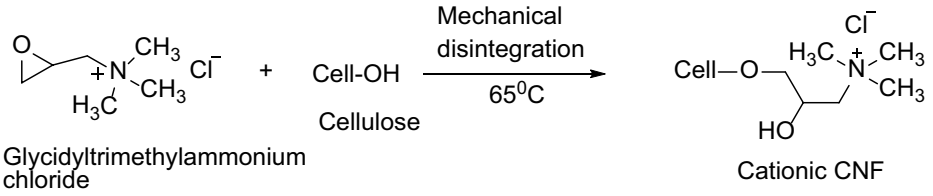
For preparation of MF, UF, NF and RO membranes various types of polymers are used such as polyamide (PA), cellulose acetate (CA), polyvinylidene fluoride (PVDF), polyethersulfone (PES), polysulfone (PSF), polyvinyl chloride (PVC), polyacrylonitrile (PAN), polyimide (PI), polyethylene glycol (PEG), poly(methacrylic acid) (PMAA), poly(arylene ether ketone) (PAEK), polyvinyl alcohol (PVA), poly(arylene ether sulfone) (PAES), polyaniline, poly(ethyleneimine) (PEI) and nanoparticles (NPs). A novel polymeric material (polyethersulfone amide) was recently highlighted in many reports [18–23].

Many composite materials of Al_2O_3, TiO_2, ZrO_2, ZnO and SiO_2 are prepared and fabricated with high crystalline phase and high porosity [24], which include TiO_2-SiO_2, TiO_2-ZrO_2 and Al_2O_3-SiC and NP inclusion composites (e.g., Ag-TiO_2, Zn-CeO_2 and zeolites) and these have shown promising outcomes for the water treatment [25,26]. The next sections will elaborate on membrane preparations, performance effects, and their applications. The primary goal of this chapter is to assess membrane performance by removing pollutants from brackish water.

2. Polymer membranes

2.1. Cellulose derived membranes

Cellulose and its derivatives are used in membrane filters extending from MF to osmosis. These cellulose-based membrane filters successfully remove viruses, germs, antibiotics, pesticides, dissolved salts, medicinal chemicals, oils, synthetic colours, and heavy metals [27]. The cellulose acetate (CA) has historic significance as one of the first compounds utilised in RO membranes. The textile dye industry effluent and a few chlorinated pesticides, including benzene hexachloride (BHC), dichloro diphenyl trichloroethane (DDT) and lindane (γ-HCH) were removed using the RO technique with CA membrane [28]. The RO with nanofilters was developed using a CA membrane and is used to treat saline water, surface water, food processing water, and industrial water and effectively removes contaminates such as ethanol, methanol, phenol [29,30], pharmaceutical active compounds [31], neutral endocrine disrupting chemicals, viruses [32] and pesticides [33]. Another cellulose-based membrane, cellulose triacetate (CTA), forward osmosis (FO) membranes, effectively remove pharmaceutical chemical substances such as naproxen, diclofenac, carbamazepine, and ibuprofen from industrial effluents, while improving the water quality index [34]. To remove residues of anions, heavy metal ions, and organic species, cellulose nitrate (CN) and CA mixed membrane filters are utilised [35,36]. The most effective membrane for removing both inorganic and organic chemicals from desalination and wastewater treatment is cellulose acetate butyrate (CAB) [37]. The contaminants are absorbed by a CN-based membrane filter, and the AC is improved by the addition of active sorbent. Low-level CN addition to polymer matrix increases features such as permeability, surface hydrophilicity, selectivity, and biofouling resistance [38]. Due to high mechanical strength, tensile strength (~7 GPa), and Young's modulus (~130 GPa), the CN has a better filtering capacity [39,40]. Sehaqui et al. [41] synthesized cationic cellulose nanofibers (CNF) for the removal of humic acid (HA) and cationic dyes from wastewater. The adsorption capacity of CNF with three unique charge components was investigated using quartz crystal microbalance of dissipation, UV spectrophotometer, and zeta potential measurement techniques. The electrostatic contact between two molecules, as well as the shape of HA, influenced HA adsorption onto CNF. The nanostructured CNF adsorbed HA upto 310 mg g^{-1} at pH = 6, a substantially higher potential than previously reported in the literature. As a result of the electrostatic interaction, cationic CNF can adsorb HA and natural organic compounds. The production of functionalized cationic CNF is depicted in Scheme 1. CN with combinations of 2,2,6,6-tetramethyl-piperidin-1-yl) oxyl oxidation, cationization, enzymatic phosphorylation,



Scheme 1. Synthesis of functionalized cationic CNF.

and other processes increases the AC of nitrates, metal ions, humic acid, and colours from industrial waste [42,43].

2.2. Chitin derived membranes

Chitin with chitosan (CS) has been widely employed in the fabrication of adsorbents composite membranes to remove dyes and heavy hazardous metal ions in flakes, beads, powders and sheets. The feed is fed through a resin-filled packed column that diffuses through internal pores. These pores allow flawless filtration through composite membrane column [44]. Hybrid adsorptive membranes are more effective at removing pollutants from aqueous media than regular polymer membranes. The solutes flow swiftly as molecular diffusion membranes are substituted with high filtration rated membranes [45]. The challenge with final water collection after treatment was separation of adsorbents. Researchers used a combination of adsorption and composite hybrid membrane technology to overcome the restrictions they encountered.

The goal of the adsorptive membrane technique was to access functional groups selectively $–SO_3H$, –COOH, and $–NH_2$ from the mixture through exterior and internal membrane, similar to membrane chromatography/affinity membrane. After saturation, the membrane was regenerated and reused by removing adsorbates. The adsorptive membranes are well-known for their ease of use with pre- and post-treatment procedures, high AC, ease of stacking and scaling-up, faster diffusion, and reuse, all of which contribute to increased acceptability and commercialization [46]. Chitosan was regarded an excellent biopolymer modifier for membranes owing to its absorptive ability. To promote dye adsorption, additives were added to chitosan with the primary goal of modifying membrane surface charge to improve dye adsorption by chemical bonding or electrostatic interaction [47]. Applying the similar strategy, Eriochrome blue black R and Alizarin dyes were adsorbed on chitosan [48]. Scheme 2 demonstrates potential H-bonding and electrostatic interactions for the adsorption of Direct blue 71 on chitosan.

2.3. Starch derived membranes

Starch is a widespread and abundant biopolymer found in plants as an energy storage element. For diverse applications, starch is combined with amylopectin, polyglucans, and amylose, a kind of glucose. Still, starch has limitations such as a high tendency for deterioration, thermal decomposition, and low shear resistance [49]. Many experiments have been reported to introduce different groups into starch for adsorption of various metals to overcome the constraints [50]. Few studies show that starch is the most effective way to remove meal ions, pigments, and phenolic chemicals [51]. The starch has the remarkable feature of self-creating porosity by sintering at temperatures ranging from 300°C to 600°C [52].

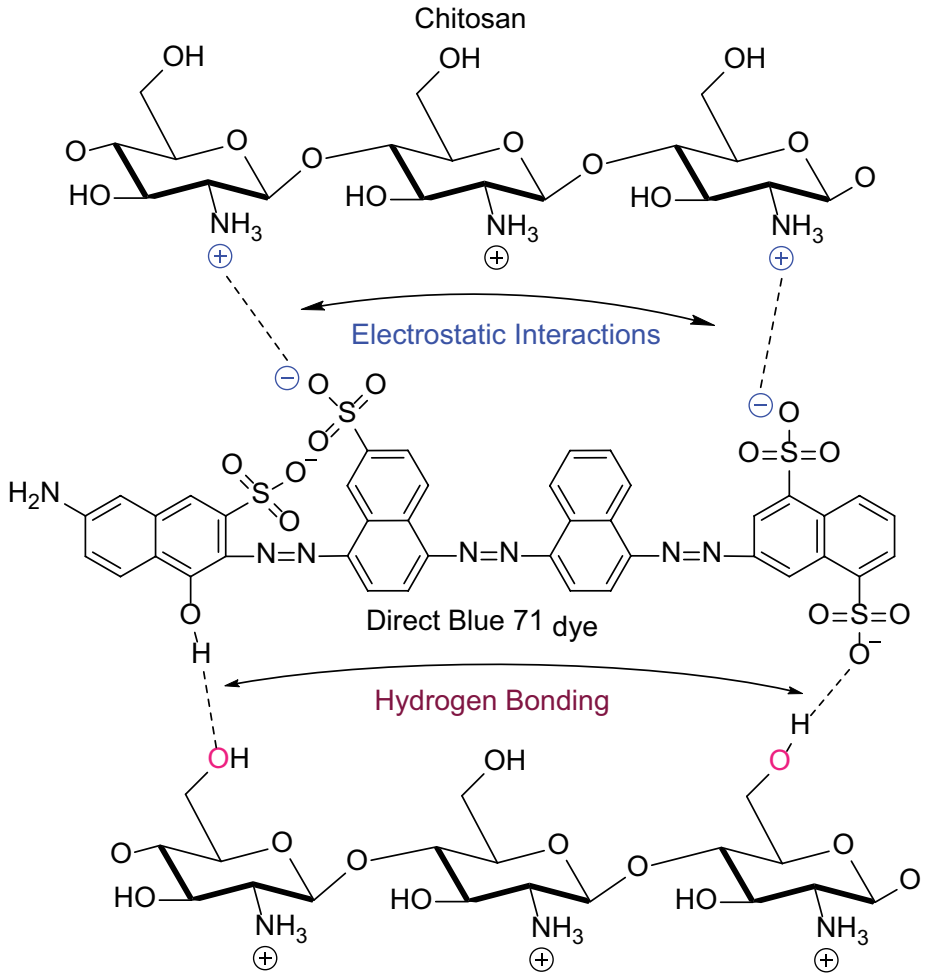
The effect of particle size and impurity of diverse starches over distinct MF membranes with combinations of precursors such as, albite, ceramic matrix made with mullite and quartz were investigated [53]. Porosity and permeability coefficients were found to be linearly proportional to the starch particle size. A recent study discovered that a starch derivative cyclodextrin (CD) can also be utilized to clean sea water. Water diffusion from pores was accounted for the CDs performance and accessing the functional groups on the surface improved membrane permeability and hydrophilicity. The CDs are cyclic oligosaccharides comprising 6/7/8 glucopyranoside units and a molecular diameter of several angstroms. CDs was synthesised through enzymatic process including CD glycosyltransferases (CGTases) and α-amylases [54]. The ironic –OH groups in the structure resulted more hydrophilic nature [55]. To enhance the porosity hydrophilicity, several ways were explored and chemically altered. CD was functionalized for primary –OH to amine groups, sulfonyl groups, and other groups to improve the membranes [56]. Due to interconnected holes in filtering, the porosity of CDs membranes demonstrated high permeability [57,58]. CD was utilised as an additive because of its poor mechanical qualities, which reduces the efficiency of application [50]. CDs, like any other polymer, are not recommended in membrane technology until novel qualities are identified.

3. Blended membranes

It is required to develop novel materials that are blended with polymer-based materials in order to improve the pure component's characteristic qualities.

3.1. Cellulose supported membrane

Various approaches have been utilised to blend pure cellulose or cellulose derivatives with natural or synthetic polymers to enhance the properties. The increased attributes improved swelling capability, selectivity, and flux. These blended membranes effectively remove hazardous components, colour as well employed in the desalination process and protein adsorption [59–63]. A phase inversion process was employed to synthesize a CA membrane supported PVC blend using N-methyl-2-pyrrolidone (NMP) as a solvent and PEG as a pore forming. The blended CA/PVC increased mechanical strength and during synthesis increase in CA content reinforced the spongy structure [61]. According to



Scheme 2. Proposed hydrogen bonding electrostatic interactions between chitosan chain and Direct blue 71 dye [48].

the report, 3% CA blending led 98%–99% salt rejection. The flow diagram of the unit utilized to check the efficiency is shown in Fig. 1.

Puspasari et al. [62] combined PEI and trimethylsilyl cellulose (cellulose precursor-TMSC) with NF and found it to be very successful in terms of cost and toxicity. This blending took advantage of a green process known as regeneration of cellulose (RC) via acidic vapor-phase hydrolysis (VPH). The filtration properties of composite RC-PEI membranes were examined and compared to those of individual RC and PEI membranes. To optimise the performance, feed PEG with blending dyes (negatively charged) was employed at a concentration of 1 g L^{-1}. The synthetic membranes made from pure RC and pure PEI had exceptional structural qualities and were tightly bonded to the top layer of PAN support.

Pure RC had a stable water flux of 6 L m^{-2} h^{-1} (LMH) at 4.5 bar feed pressure with 80% dismissal of PEG with 400 Da, but pure PEI had a superior water flux of 8 LMH with less dismissal of PEG. The RC-PEI blend led to the compound (400 Da mol wt) with more PEG resistance while keeping the same water flux as pure. Physical properties of cellulose blended polymers like as hydrophilicity and permeability affected the membrane's mechanical strength. The increased permeability and hydrophilicity reduced mechanical strength. CA supported hydrophobic polymer blends were shown to be more effective in changing the properties of AC of the water flux and structure.

Riaz et al. [63] introduced the CA supported polyurethane (PU) blended membrane to treat textile effluents. The CA:PU were prepared in different ratios, that is, 15:85, 20:80, 25:75, and 30:70 in the presence of tetrahydrofuran (THF). To expand the chemical resistance CA in PU blending, the CA:PU ratio mixture was preheated in a Teflon walled container and then evaporated at 55°C. Scheme 3 shows the physical bonding between CA and PU. Another CA/PU blend (20% PU and 5% CA) in acetone and dimethylformamide (DMF) (7:8 mass ratio) solvent mixture, was found to be stable in pH 4~10, but pure CA was unstable on the higher end of the pH range. Apart from various treatments, the aqueous media of textile dyes were treated with sodium-2-((2-hydroxy-6-sulfonatonaphthalene-1-yl) diazenyl)-6-methoxybenzothiazole-5,7-sulfonate) and its derivatives containing Fe^{3+} ions complex. The complexed dye filtration was 10% higher than the pure CA membrane [64].

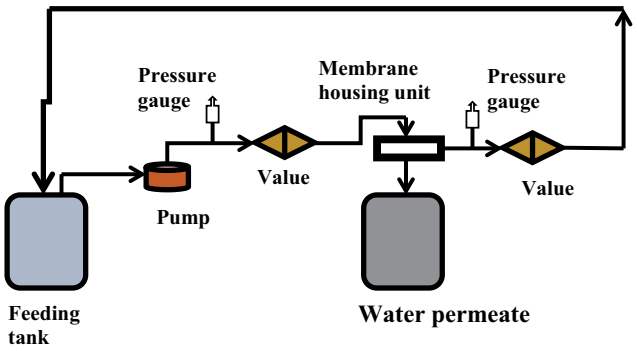
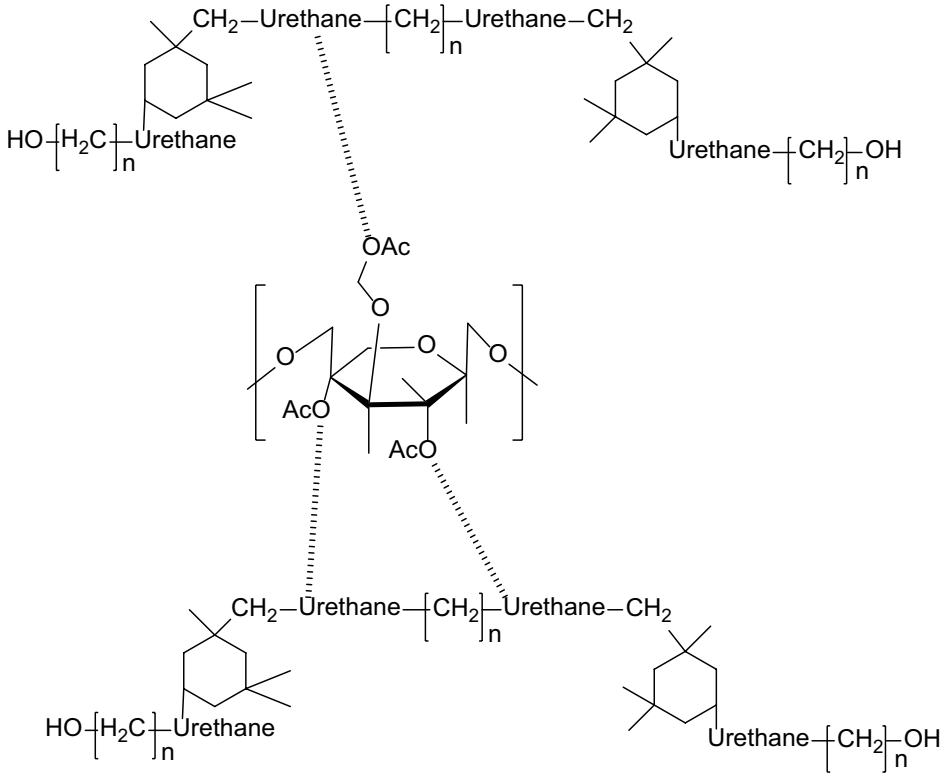


Fig. 1. Laboratory unit for removing salt [60].

CA supported PES membrane with a thin outer layer possessed big pores on the surface. The CA/PES blended membrane was synthesized using the immersion precipitation process, which resulted in reduced selectivity and a 10% oil separation ability, compared to pure PES membrane [65]. Pure cellulose has also been combined with a number of biopolymers to generate completely functional bio-based composite materials, such as chitosan-cellulose blends. Chitosan was used to make a variety of useful materials, including fibres, sponges, nano-whiskers, and beads.

Many studies on composite blends revealed superior physical properties such as self-healing [66], antibacterial [67], higher surface porosity and interconnected void structures [68], potential antistatic and moisture absorption characteristics [69], adsorption of metal ions, upgraded water absorption capacity, and high mechanical properties [70]. Chitin/cellulose was prepared by freezing or thawing a mixture of NaOH (7 wt.%) and urea (12 wt.%) in an anionic solvent, which was regenerated at the end of the process. The porosity of this chitin/cellulose membrane ranged from 100 to 800 nm, which improved adsorption's efficacy in removing dangerous metals like Cu^{2+}, Hg^{2+}, and Pb^{2+}. The strong adsorption efficacy was attributed to the interaction of chitin –OH groups. As the amount of chitin in the cellulose mix increased, the AC of metal ions improved. The amount of Hg^{2+}, Cu^{2+} and Pb^{2+} adsorbed over the unit amount of bended membrane was found to be 3.85, 2.00 and 2.44 mmol g^{-1}, respectively [71].

Cellulose and its derivatives are also combined to generate membranes that are both environmentally benign and long-lasting, such as CA/CTA and CA/CMC. Forward osmosis (FO) was evaluated on CA/CTA composite membranes, whereas UF was studied on CA/CMC acetate composite membranes, with noticeable features where pure membranes fell short. Over thin-film composite (TFC) membranes, CA/CTA composite membranes utilising the FO technique have been commercialised for chlorine resistance and biodegradability [72]. Chen and Lee [73], also confirmed that metal ion removal parameters for CA/CTA composite membranes employing the FO technique were satisfactory. The immersion precipitation procedure was used to create a membrane using a mixture of 1,4-dioxane, acetic acid, and acetone. Fake wastewater was created in a 1 M NaCl solution containing 100 mg L^{-1} of Zn^{2+}, Ca^{2+}, Cu^{2+}, Mg^{2+}, Hg^{2+}, Cr^{2+} and Pb^{2+} ions to test the membranes



Scheme 3. Physical bonding between CA and PU [63].

filtering capacity. This process was found to be significant because of its ability to eliminate hazardous metal ions, which is comparable to commercial goods (Hydration Technology Innovations, HTI-TFC). The setup employed for FO process is shown in Fig. 2.

Han et al. [74] developed CA blended carboxymethyl cellulose acetate (CMCA) membrane employing phase inversion PI with the incorporation of PEG-600. The membrane showed promising hydrophilicity as carboxylmethyl functional groups altered water contact angle. The absence of intermolecular bonds in blended polymers reduced thermostability. Furthermore, the hydrophilic nature of the membrane promoted anti-fouling capability, as well as the ease of processing and cost-effectiveness. Finally, combining cellulose with the proper polymer is the only practical technique to develop a high mechanical strength, water permeable and hydrophilic membrane with stability.

3.2. Chitin supported blended membranes (BMs)

Chemical stability and mechanical strength concerns were triggered due to CS coating over polymeric membranes. These CS composite membranes featured a few flaws, such as non-uniform coating and defects in the CS film during manufacture. Blending was utilised as a one-step procedure in bulk production. Blending with CS increased performance by acting as a stabilising and chelating agent. Furthermore, decreasing the CS concentration in the membrane matrix resulted in porosity with poor swelling efficiency [75]. PES is a prominent polymer additive, however, its hydrophobic properties limit its usage. The crosslinking agent glutaraldehyde (GA) solution was used to combine acrylonitrile butadiene styrene (ANBS) over CS and PES for manufacture to filter Na^{+} and Hg^{2+} ions. The XRD patterns of this polymer blend showed that addition of CS enhanced the porosity and amorphous phase of the membranes. The amorphous phase saturated flux, causing swelling to grow. Furthermore, selective CS coating provides desirable permeation rate and hydrophilicity.

PSF/N-succinyl-chitosan (PSF/NSCS), PSF/CS, PSF/N-propyl phosphonyl chitosan (PSF/NPPCS), and polyethyleneimine (PEI) blends were made using UF membranes for heavy metal ion adsorption. Scheme 4 depicts a general complexation process of PEI with metal ions. The immersion precipitation process is used to create these blends prior to mixing. For selective removal of metal ions, large amounts of CS were applied to the PSF matrix. NPPCS and NSCS polymers were strong enough to remain in the column after lengthy runs and exhibited high solubility in the PSF matrix. To remain stable through immobilisation,

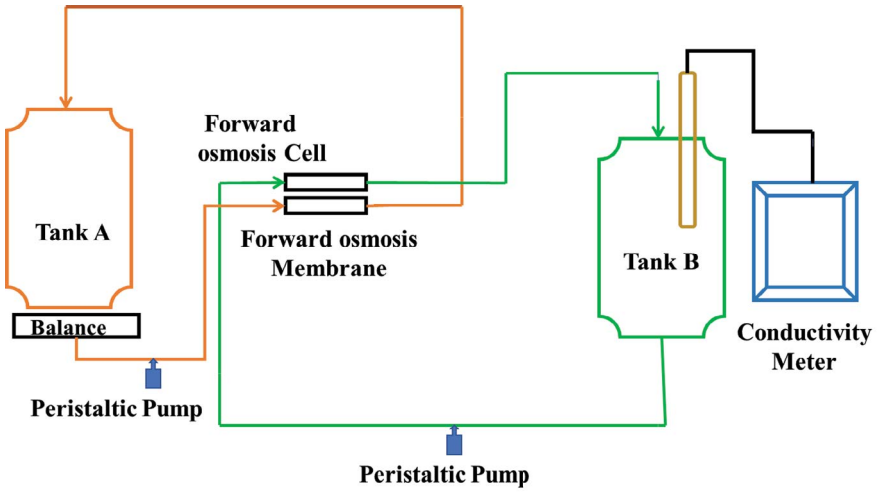
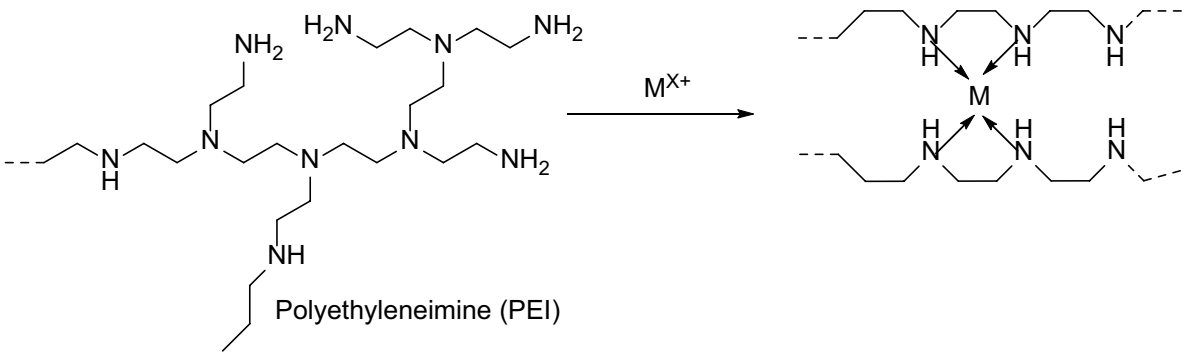


Fig. 2. Flow chart for the forward osmosis process (Tank A = Feed tank, Tank B = Draw tank) [73].



Scheme 4. Complexation of PEI and metal ions [76].

NPPCS and NSCS polymers were physically adsorbed and crossed linked with matrix. In order to prevent fouling, the PSF matrix was mixed with CS, NSCS, and NPPCS to generate a hydrophilic membrane [76]. The active and specific surface area is highly demanded for adsorptive purposes by combination of spinning and coagulation methods.

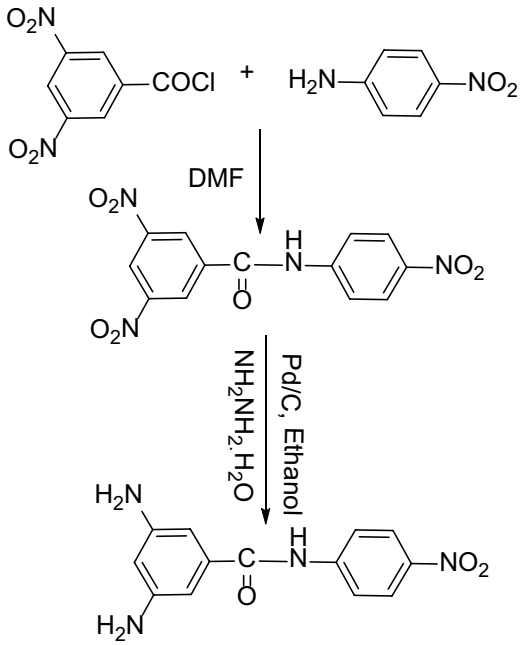
4. Composite/mixed matrix membranes

The mixed matrix membranes (MMM) are made up of both organic and inorganic compounds, where inorganic substances are distributed in whole polymer matrix membranes as fillers. These fillers significantly improve the characteristics of the polymer matrix. The molecular sieve like fillers present in small amounts in polymeric matrix showed outstanding increase in the selective characteristics of the new material in polymer matrix [77]. Many studies were carried out to alter the physical properties of cellulose-based MMMs with various kinds of fillers. For the purposes of industrial research, the MMMs were doped with NP fillers and were known as polymer-nanocomposite membranes which led to substantial improvement.

4.1. TFC membranes

Thin films have been presented as a solution to thick membrane limitations such as low flux and high selectivity. Porous membranes have a high permeability and limited selectivity. TFCs are made in a two-step process. The ultra-thin layer was applied on a thick and a porous non-selective substrate. Interfacial polymerization (IP) [78,79] and dip-coating [80,81] were used to create a stable top layer. IP is quite popular as two monomers react with each other for which immiscible solvents act as an interface. IP resulted in the formation of extremely cross-linked polymer under controlled conditions such as ultraviolet (UV), heat, and plasma treatment. The produced TFCs were remarkably thin (≥0.1) with great permeability.

The effect of pure and altered PVC membranes over TFC layers was practiced for desalination process. The alteration of the PVC with (3-aminopropyl)triethoxysilane (APTES) was effectively used. The PVC supports prepared with various compositions of APTES from 0–6 wt.%, showed noticeably change in crystal morphology with surface characteristics. The modifications were fabricated with PA through identical IP by cross-linking of the polymer structure. The IP technique parameters of flux for TFC membranes was 0.89 LMH bar^{-1} and modified PVC support layers was 2.70 LMH bar^{-1} [82]. In another study, the spongy structure was created by mixing sulphonated poly-(etherketone) (SPEK) with TFC-FO membrane substrate to achieve high hydrophilicity. Another experiment used the monomers 1,3-phenylene diamine (MPDA) and 1,3,5-benzenetricarbonyl trichloride to create a thin and selective layer using the IP technique. TFC-FO membranes combined with 50 wt.% SPEK usually resulted in a thin, spongy structure with fluxes of 50 and 35 LMH [83]. The amine monomer β-cyclodextrin polyurethane, that is, DABA (Scheme 5) was blended with MPDA/TFCs/PAN to increase the hydrophilicity of each component. The high



Scheme 5. Formation of DABA [84].

DABA content significantly increased water flux from 37.5 to 55.4 LMH with 98% of high salt rejection and antifouling property [84].

The efficiency of foul resistance with BPA and/or TMBPA was confirmed over TMCIP. Low fouling was due to high negatively charged polyester layer embedded on the top of TFC composite membrane [85]. To recover the antifouling, PVA was deposited on NF/TMC composite membrane mixed with piperazine by IP. The incorporation of a hydrophilic PVA chain into the active layer of PA resulted in a smoother surface and better hydrophilicity and the elimination of fouling during extended operations [86]. TFCs were also modified with hydrophilic additives (macromolecules) for tunable performance of membrane [87,88]. Acetone as a solvent was added to polymeric phase for preparing TFC/NF membranes with active layer of nanosized porous structures [89].

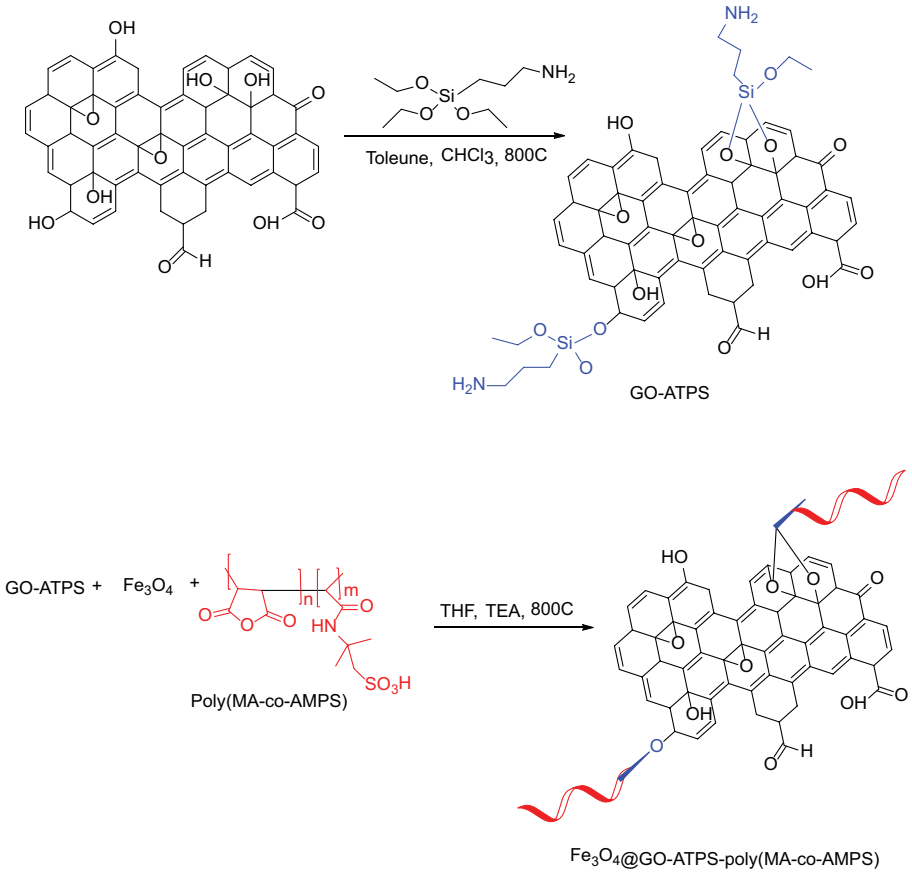
4.2. Cellulose supported MMMs

MMMs are well practiced due to its promising scaled-up separation capabilities. Tian et al. [90] found promising adsorption of Rhodamine-B and organic based dyes after the addition of graphene oxide (GO) to cellulose matrix. Cellulose-GO membrane was synthesized through vacuum-filtration with ultra-sonification. The PVDF film was used to pass the filtrate for better casting dope. The cellulose-GO composite membrane (mass ratio of 8:1) demonstrated promising 86.4 mg g^{-1} AC, as compared to pure cellulose with 0.6 mg g^{-1} AC. Cellulose-GO composite membrane showed improved AC which was accounted by high amounts of oxygen-containing functional groups and robust the intermolecular interactions. These tough intermolecular interactions unveiled a well dispensability and thermally stability in both GO and cellulose MMMs.

Scientists have made many attempts to elevate the hydrophilic character as it is crucial for filtration process through any membrane. Sahraei et al. [91], tested the loading of Fe_3O_4 (0.5 wt.%) over GO-(3-aminopropyl) triethoxysilane-2-acrylamide-2-methyl propanesulfonic acid-maleic anhydride NC (Scheme 6) for effectiveness of thermal stability of CA.

Colburn et al. [92] verified the changes in cellulose membrane's physical properties with selective loading of graphene quantum dots (GQDs). At the initial phase cellulose/GQDs were processed using ionic liquid (ethyl-3-methylimidazolium) for effective solubility of cellulose. The casting dope was immersed into isopropanol or water for gelation. The gelation with resilient was due to the –COOH and –OH groups in GQDs and enabled the interactions in membrane. The high performance of cellulose/GQDs were due to the presence of oxygen containing functional groups leading to substantial improvement in characteristics like level of hydrophilicity and permeability of water as compared to unmodified membrane. Parallel observations were also reported with TiO_2 NP coated over CA supported matrix and it was found that, with the increase of TiO_2 content, the stability of membrane improved. TiO_2/CA stability was due to close-fitting of covalent bonds amongst TiO_2 filler and CA matrix, which resulted in the effective firmness of the polymer chain [93]. Rajesha et al. [94] reported the use of diffusion induced phase separation technique (DIPS) to load ZnO/zeolite over the CA supported polymer matrix demonstrating the refusal properties. The benzophenone-3 (BP-3) solution was used in the filtration process. The 98% of BP-3 was eliminated with high amounts of ZnO in zeolite support at 200 kPa over MMM.

Mukherjee and De [95] observed the effect of Al (from 10 to 25 wt.%) NPs by immersing into cellulose acetate phthalate (CAP), which has an abundance of –OH and –CO functional groups. This MMM synthesis was carried out by PI technique using solvent of DMF. The increased Al content from 0 to 20 wt.% effectively increased the porosity of the membranes. The advantage of selective Al addition to matrix was the separation of polymer chains and sustained with high stability. The MMM porosity enhanced the AC of derivative compounds of phenol like para-nitrophenol (PNP), meta-nitrophenol (MNP), catechol (CC), ortho-chlorophenol (OCP) at 9.0 pH and 100 ppm. The MMM porosity showed the adsorption and rejection order of CC > PNP > OCP > phenol > MNP. Many reports confirmed that properties like high filtration, strength, selectivity and antifouling can be obtained by dispersing inorganic fillers in cellulose based polymeric matrix [96]. The researcher's main goal is to find a high-performance filler as amount of filler loading is challenging, and it ultimately lowers the mechanical stability [97].



Scheme 6. Synthesis of Fe_3O_4 (0.5 wt.%) over GO-(3-aminopropyl) triethoxysilane-2-acrylamide-2-methyl propanesulfonic acid-maleic anhydride [91].

4.3. Chitin supported MMMs

MMM offers better properties (mechanical, chemical, and thermal) by integrating various types of fillers into polymer matrix. The selective loading of compatible NPs results in increase of hydrophilicity, permeability, and mechanical strength. Mukhopadhyay et al. [98], improved rejection of As^{3+} ions by addition of zeolite particles (0.25–1.25 wt.%) over CS membrane with the feed strength of 500–1000 μg L^{-1}. The 1.25 wt.% zeolite demonstrated more rejection at 31.64 kgf cm^{-2} pressure. The zeolite particles were ultrasonicated with mixture of glacial acetic acid and CS mixture in various ratios. Using the dry PI process, the resulting mixture was placed layer by layer on non-woven cloth. Like any other membranes, this composite zeolite/CS provides a hydrophilic environment along with properties like soft surface, antifouling and greater water flux. Shao et al. [99], confirmed the improved performance by GO incorporation over CS membrane, prepared by drying process with acetic acid.

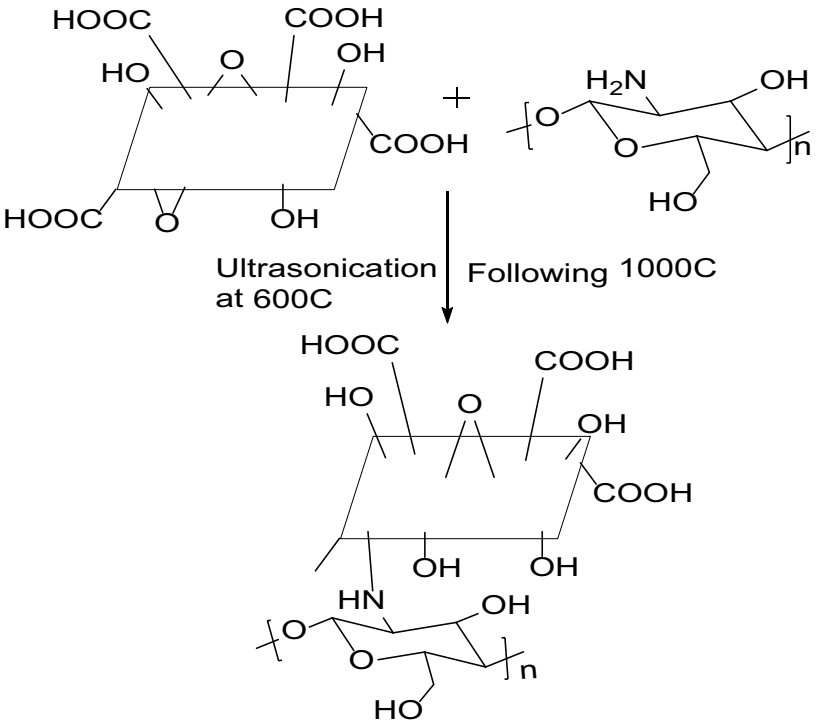
The notable performance was obtained by 1 wt.% GO addition to NC membrane which leads to high tensile strength of 104.2 MPa, compared to 4.32 MPa of pure CS membrane. This is because amino acid of CS and epoxy of GO were crosslinked (Scheme 7). This crosslinking provided the unvarying, well dispersed GO covered matrix with robust surface interface resulting in tough properties.

The complete bio-NC membrane over CS matrix was prepared by Karim et al. [100] through freeze drying technique using cross-linking agent as a GA vapors followed by compacting. The bio-NC/CS membrane was meant for cationic dyes adsorptive filtration. Adsorption ability and mechanical strength were significantly improved after adding cellulosic NCs to the CS matrix, whereas pore size, flux, and surface area were somewhat reduced. The bio-NC/CS membrane showed greater affinity to eliminate the dyes, among methyl violet, Victoria blue and Rhodamine 6G. The first mechanism of dye adsorption is through hydrogen bonding and second is through interface between dyes with positive charge and cellulosic NCs with negative charge (Scheme 8). A few uncross-linked polymer matrixes were under swelling, still their efficiency remained intact.

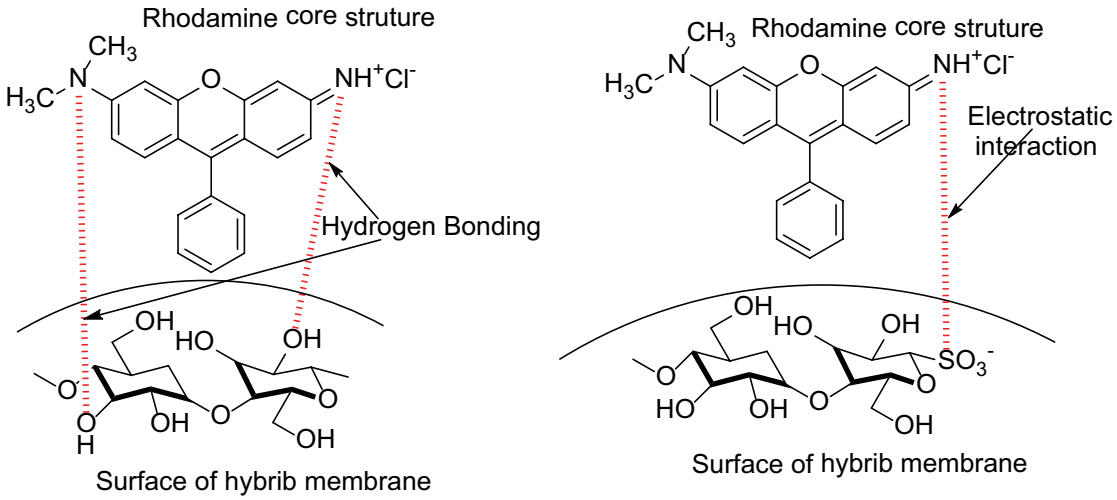
Munnawar et al. [101], prepared CS-zinc oxide hybrid NPs (CS-ZnO/HNP) to combined PES matrix in various concentrations. This CS-ZnO/HNP/PES composite is prepared by PI technique via water coagulation bath for antibacterial and antifungal activity. This remarkable antibacterial and antifungal activity are due to synergistic effect of CS and ZnO to counter the bacteria, that is, *B. cereus*, *S. aureus*, *E. coli* and fungi, that is, *F. solani*, *A. fumigatus*. The CS-ZnO/HNP blend provides flatter surface and boosted hydrophilic character of the MMMs.

5. NC membranes/nanofibrous (NF) membranes

In order to upgrade the filtration performance researchers synthesized various NPs doped polymeric materials which are addressed as NC polymeric membranes, these have enhanced physicochemical properties. Apart from NPs, nanomaterials such as nanoclay, carbon nanotubes, nanosilver, nanosized materials containing ZnO, TiO_2, SiO_2, Al_2O_3, Fe_3O_4 and ZrO_2 are developed to produce high performance NC membranes for desalination process.



Scheme 7. Cross-linking reaction occurring during the preparation of NC membrane [99].



Scheme 8. Binding mechanism of dyes with cellulosic NCs [100].

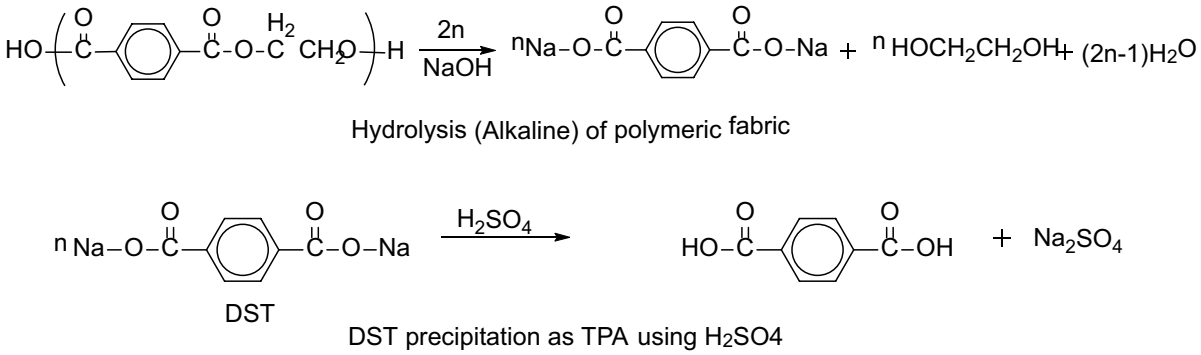
5.1. Metal and metal oxide NC membranes

The usage of metal nano-particles in polymeric membranes as composite took a hype, due to its tuneable significant characters like strength, hydrophilicity, permeability, and selectivity in seawater treatment. For example, addition of ZnO in CS membrane provides high antibacterial activity and strength [102], silica NPs into PVDF membranes increased the selectivity, temperature resistance, and diffusivity with a remarkable difference [103], on adding aluminium oxide [104] and silica NPs (using membrane bioreactors) [105] into PSF membranes, increased the permeability of gas and porosity with low flux. The addition of silica NPs on polybenzimidazole membrane leads to higher selectivity, permeability, and gas separation [106]. Apart from the above tuneable characteristics, metal oxides as NPs has few limitations. For example, addition of TiO_2 more than 2% in PSF membrane led to decrease in permeability [107]; metal oxides in higher quantity caused negative effect by enlarging pore size which disturbed the selectivity and permeability; risk on health and environmental also a big concern. The synthesis of membranes with integrated NPs as functionality material were intended to disperse the metal particles with less than 100 nm in diameter. Choe et al. [108] employed SelRo (MPT-34) nanofiltration membranes with membrane areas of 0.024 and 5.2 m^2 and recovery of NaOH and disodium terephthalate (DST) from alkali trash of polyester fabrics were examined, respectively. Volumetric concentration factor (VCF), filtration time, temperature, operating pressure, and cleaning flux behaviours were detected. Presence of ethylene glycol (EG), NaOH and DST were calculated using various VCF values. The NF process resulted in a concentrated DST solution that required less acid to precipitate and a NaOH permeate comprising EG and DST. The concentrations of EG and DST that control permeate reusability in the NaOH treatment of polyester fabrics were found to be VCF dependent. The most appropriate VCF for retaining relatively decent membrane production and high DST rejection was around 4.5. DST and EG rejections were 86% and 42% at a VCF of 4.5, respectively, whereas NaOH recovery was 84% with fluxes between 12 and 36 LMH. For alkali wastewater from polyester materials, many wastewater remedies have been employed. To precipitate DST as terephthalic acid (TPA), it is commonly acidified with H_2SO_4 to a pH of 2–3, as indicated in Scheme 9. Hong et al. [109] improved the agglomeration by addition of metal NPs. Gilbert et al. [110], observed that agglomerations of NPs were persuaded by increasing ionic strength and pH.

5.2. Cellulose-based nanofibrous (NF) membranes

For treating brackish water, typical polymeric membrane modifications had a few drawbacks, such as inadequate void size with dispersion, suboptimal water flow, less porosity, and a high fouling propensity [111]. Many researchers are focused on the improvement of the characteristics to overcome the limitations in the traditional process [112]. The electro spun cellulose and its derivatives provide high hydrophilicity, less poisonous, readily accessible in nature, acceptable biocompatibility, and likely functional ability in the synthesis of polymeric membranes. Hong et al. [113] prepared NF cellulosic with high porosity structure, high filtration flux, and oil pressure disturbance. Usually, NF cellulosic was prepared by wet electrospinning technique leading to well-organized stacking of porous NF membranes. The cellulose in NF cellulosic reduced the oil droplet angel in seawater by increasing the chemical stability of bovine serum albumin (BSA) and bilirubin. The RC membrane micro filters were commercialised with regeneration and reuse for up to three times [114].

Zhang et al. [115] observed CA NF mat prepared by electro-spun method with adsorptive membrane of diethylaminoethyl (DEAE) anion-exchange ligand with strong and stable interacting ability of 40.0 mg g^{-1} for BSA, whereas commercial RC membrane, cellulose microfiber medium, and cotton balls were found to be 33.5, 14.5 and 15.5 mg g^{-1}, respectively. These statistics confirmed the effectiveness of electro-spun technique for the incorporation of hybrid NF into the biopolymer matrix, leading to the changes in the surface functional groups that substantially improved



Scheme 9. Saponification of polyester fabrics and standard wastewater treatments [108].

AC. By adopting the careful and effective method of assembling, the organic-inorganic hybrids extensively improved the surface area and thermal stability as compared to pure polymer matrix accounting for the higher lipophilic and specific toxic character [116]. In a unique study the CA with NH_2-functional group was doped over silica composite NF membrane (FCA/SiO_2) by sol-gel technique and electrospinning method for the elimination of Cr^{6+} ion. The high porosity was achieved by both techniques and surface area of 126.49 and 80 $m^2 g^{-1}$ was attained by sol-gel and electro-spun of FCA/SiO_2 membrane respectively. The high surface area allowed high AC (19.46 mg g^{-1}) of Cr^{6+} ion over the FCA/SiO_2 membrane, which was much higher than α-Fe_2O_3 nanofibers (16.17 mg g^{-1}), PAN/ferrous chloride composite nanofibers (4.4 mg g^{-1}) and hematite hollow microspheres (6.8 mg g^{-1}). The high ability of adsorption was because of the abundant surface area binding the NF membrane and FCA/SiO_2 effectively [117].

5.3. Chitin supported NF membranes

Huge porosity, larger specific active surface area, and easy customization of surface functions are some of the characteristics of an ideal membrane. Electro-spun membranes are used for efficient adsorption of contaminants (organic and inorganic) during waste-water treatment. The CS-based electro-spun membranes effectively remove metal impurities such as Co^{2+}, Cr^{6+}, Ni^{2+}, Pb^{2+} and Cu^{2+}. Taha et al. [118], aimed for selective absorption of Acid blue 113 and an anionic dye by using pure CS-NF membrane which was prepared by electros-pinning and produced fibres with diameter of 86 ± 18, 114 ± 17,164 ± 28 nm. Highest adsorption of 1338 mg g^{-1} was received over fibre (86 nm diameter) due to increased surface area providing increased adsorption. The adsorption mechanism involved is illustrated in Scheme 10.

For effective synthesis of CS-NF membrane, blending method is well established due to intra- and inter-molecular connections, polycationic solution and complex chemical structure compared to mere CS nanofibers [120,121]. The revolting forces between ionic groups of the membranes were interrupted by unbroken fibre development [122]. CS-nanofibers have many setbacks such as low adsorption ability after long runs which is due to swelling of the fibres [123]. The electrospinning (Fig. 3) was enhanced by blending CS with various types of synthetic or natural polymers.

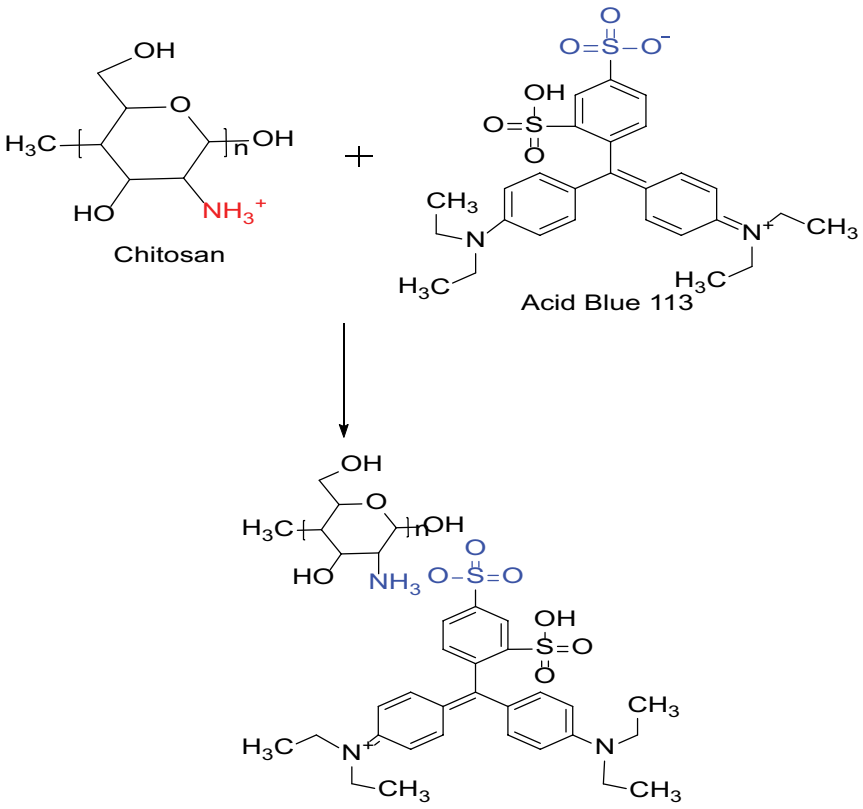
Shariful et al. [124] opted electrospinning to develop CS/PEO/AC NF membrane (CPANM) through chemical activation technique from rice husk. CPANM showed promising AC of Fe^{3+}, Cr^{6+}, Zn^{2+}, Pb^{2+} and Cu^{2+} as compared to CS/PEO fibre (CPF) due to amalgamation of AC. The tremendous adsorption processes were associated with amalgamation of AC, first one was the increased surface area (371 $m^2 g^{-1}$) and second one was addition of (–COOH) group in fibre membrane surface of CS. The bonding between CS, PEO and AC is shown in Scheme 11.

Habiba et al. [125] prepared CS/PVA blend NF membrane with high hydrophilicity with tensile power of 1.58 MPa with active specific surface area (218 $m^2 g^{-1}$). These properties provided extreme adsorption competence of Zn^{2+}, Cu^{2+} and Pb^{2+} [126]. CS/PVA blend was prepared with electrospinning method, to adsorb and remove Cd^{2+} and Pb^{2+} from the effluent. This preparation shows bead-less functional active surface which strongly combined with metals Cd^{2+} and Pb^{2+} with AC of 148.79 and 266.12 mg g^{-1} respectively. The various parameters were optimised to control, (i) the time of interaction, (ii) dosage of adsorbent, (iii) opening metal ions concentration, (iv) pH of solution used and (v) effects of contending ions for better performance.

With CS-based electro-spun NF membranes, any effectively adsorbing membrane to remove heavy metals may be manufactured by improving attributes such as (i) high porosity, (ii) customised made pore size, (iii) increased volume to surface ratio, and (iv) high adsorption efficiency. The setbacks like, (i) huge rate of swelling, (ii) comparatively less mechanical strength, and (iii) considerable lowering of AC, can be overcome by improving preparation techniques and addition of composite materials to membranes [127]. Table 1 illustrates the performance of various polymeric membranes.

5.4. Starch supported NF membranes

These CDs are modified with PA TFC membranes to improve the hydrophilicity, water flux, rejection capacity with lower swelling rate. Wu et al. [145] tested physical properties of NF/β-CD/polyester TFC membranes through IP. Triethanolamine (TEOA) and trimesoyl chloride (TMC) were polymerized with the β-CD and shown in Scheme 12.



Scheme 10. Adsorption mechanism between Acid blue 113 and CS [119].

The accumulation of β-CD was led to substantial improvement in hydrophilicity, anti-fouling, and permeability with promising rejection of Na_2SO_4 compared without β-CD addition.

Similar observation was notice over TFC/*m*-phenyldiamine (MPDA)/TMC membrane, prepared though polymerization and amine-functionalized β-CD (amine f-CD) addition by *in-situ*. The flux rate was improved over modified membrane by 20 LMH, whereas unmodified membrane showed 5.59 LMH at 100 psi. Effective insertion of amine f-CDs led to the modification of the membrane to possess high surface area, hydrophilicity and porosity [146]. A few studies showed that incorporation of different CDs (diethylamino-α-CDs, amino-α-CDs and diethylamino-β-CDs) into PA-TFC membranes led to the modification of functions by increasing the crystallinity, rejection of high divalent salt and antifouling at various pH levels [147]. CDs with toroidal shape are the potential additive to remove selective organic compounds by host-guest inclusion complexation technique. This technique encapsulates different types of organic molecules such as trichloroethylene [148], pesticides [149], hydrocarbons [150] and NOM (humic acid) [56] from seawater. Adams et al. [56], tried to complete elimination of NOM (68%) by PSF/β-CD PUMMMs through immersion precipitation technique. This technique notified good hydrophilicity and water flux on addition of β-CDPU into PSF MMMs. Addition to this β-CDPU provided –OH and $–NH_2$ groups, which established the anti-fouling properties even for long water runs. To remove, di-(2-ethylhexyl) phthalate (DEHP) from portable water PSF/β-CD hollow fibre membranes were employed. The amount of β-CD effectively eliminated the DEHP (70%) from solution [151].

5.5. Graphene oxide functtionalized membranes

Desalination applications utilising graphene oxide (GO)-assisted membranes have advanced significantly, since GO functionalized membranes are more efficient and consume less energy during the desalination process. Furthermore, the GO-functionalized membranes are more chlorine resistant while maintaining the same reverse osmosis performance. Also, surface modification uses a little amount of GO nanomaterial, enabling it non-expensive and reduces the environmental impact caused by the GO production [152,153].

Zhang et al. [154] used the layer-by-layer self-assembly (LBL-SA) approach to deposit GO nanosheets followed by amino-GO (aGO) nanosheets upon that surface of amino poly ethersulphone (aPES) membrane in an early experiment to manufacture GO-surface modified membrane. The produced GO/aGO/aPES reverse osmosis membrane is significantly more chlorine resistant than the initial polyamide reverse osmosis (PARO) membrane, it also has excellent salt rejection rate of upto 98% and water flux of 28 L m^{-2} h^{-1}. On the aPA-TFC membrane, the same modification scheme was used; the LbL-SA approach was also used to deposit GO's and aGO's multilayers on the surface

Table 1
Various polymeric membranes with % of salt rejection and flux

S. No.	Type of membrane	Rejection of salt	Flux	References
1	PVC/CA	99.9% salt	40 LMH	[61]
2	CA/PU	97.4% BSA	13.93 LMH	[63]
3	CA/CTA	99.5% NaCl	9.27 LMH	[72]
4	PSF/N-succinyl-chitosan	98% Cu^{2+} 95% Ni^{2+} 92% Cd^{2+}	117 LMH	[76]
5	GQD/cellulose	95.9% PEG 80% MB	20 LMH	[92]
6	Cellulose acetate phthalate/Al	91% catechol	42 LMH	[95]
7	CA/PEG/SiO_2	92% salt	2.09 LMH	[97]
8	Zeolite/CS	>94% As^{3+}	37.08 LMH	[98]
9	Cellulose	99% oil	12,000 LMH	[113]
10	CA/PSF	84% BSA 70% Cu^{2+}	47.5 LMH	[128]
11	PVDF/CA	45.2% BSA	240 LMH	[129]
12	CS/CA	81.03% Cu^{2+}	309.2 LMH	[130]
13	CA/GO	90% salt	65 LMH	[131]
14	Polyethylene terephthalate/PVA/RC	>90% oil	12.7 LMH	[132]
15	Polyethylene terephthalate/PES/RC	>90% oil	10.7 LMH	[132]
16	CA/PMMA	99.9% humic acid	334.5 LMH	[133]
17	Polyethylene terephthalate/PAN/cellulose nanofiber	99.9% oil	208 LMH	[134]
18	PVDF/cellulose nanocrystal	>97% toluene >95% oil	5,842 LMH	[135]
19	Polyethylene terephthalate/cellulose nanocrystal	99.5% oil	272.3 LMH	[136]
20	PEG/chitin NPs	>99% oil	217 LMH	[136]
21	PES/cellulose nanocrystal	97% BSA	168 LMH	[137]
22	Ceramic/CS	100% Cu^{2+} (approx.)	5–50 LMH	[138]
23	PVDF/nanoclay-CS	>95% Acid orange 7	220 kg m^2 h^{-1}	[139]
24	PSF/CS	97.2% BSA	260 LMH	[140]
25	PES/GO/CS	>90% Na_2SO_4	41 LMH	[141]
26	Carbon molecular sieve-Al_2O_3	>93% NaCl	25 kg m^2 h^{-1}	[142]
27	PVDF/SiO_2 NPs	>99.9% NaCl	30.7 LMH	[143]
28	PSF/PTFE	99.99% NaCl	39.5 LMH	[144]

of the membrane. While retaining separation performance, a double-functionalised protecting layer was fabricated to enhance the chlorine resistance and membrane anti-fouling capabilities. On comparing with the unmodified membrane, salt rejection was reduced by 0.7% and water flux was enhanced by 10% [155].

Recently, researchers have focused on integrating GO into polymeric mixed solutions, which are then combined to boost water permeability, mechanical strength, and antibacterial properties [154,156–159]. Before being integrated into a polymer matrix, GO can be pre-functionalized, or it can be applied without being pre-functionalized. Various modifiers such as hyper-branched polyethylenimine (HPEI), isocyanate (I) [160] and 3-aminopropyltriethoxysilane (APTS) [161] were used to modify GO. After that, various proportions of functionalized graphene oxide were combined with polysulfone (PSF), PVDF polymeric solution, respectively to be casted using conventional phase inversion process. The developed ultra-filtration membranes I-GO/PSF and APTS-GO/PVDF were able to decline BSA by 95% and 57%, respectively, whilst the HPEI-GO/PSF declined the polyvinyl alcohol (30,000–70,000) upto 90% and polyethylene glycol-20,000 up to 85%. The mechanical characteristics (elongation and tensile power) of APTS-GO/PVDF membranes were enhanced by 48.38% and 69.01%, respectively, when compared to GO/PVDF membranes due to the strong inter facial interaction between the polymer matrix and APTS-GO. The reaction between APTS and GO is depicted in Scheme 13.

6. Future perspectives

Communities are resorting to desalination as the ideal approach for ensuring consistent water supply in the face

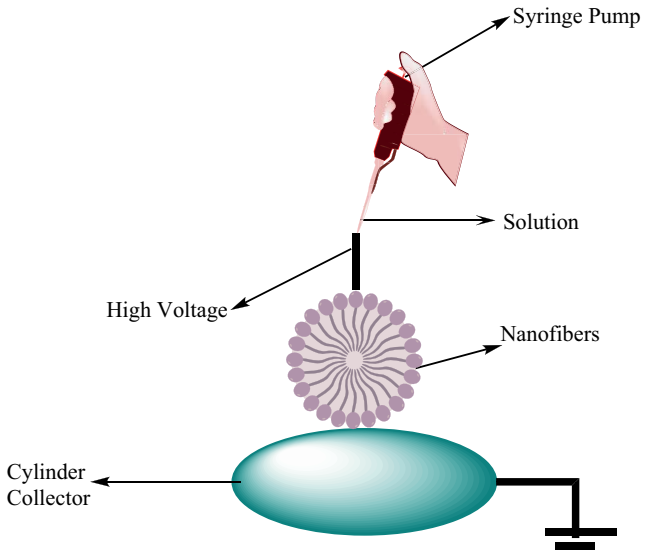
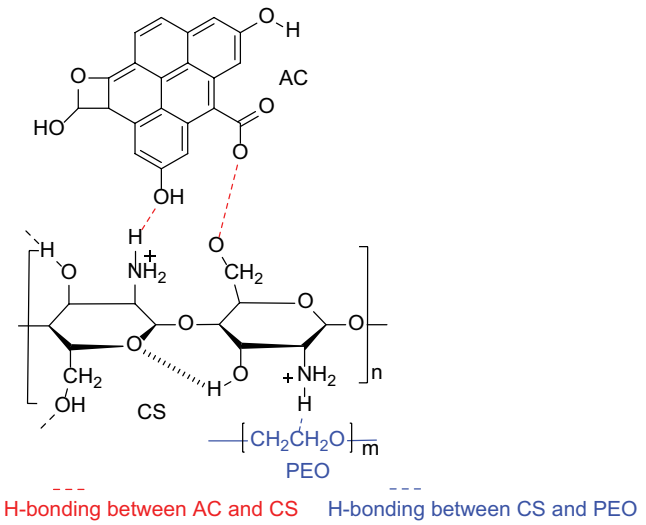


Fig. 3. Electrospinning process [123].



Scheme 11. Proposed H-bonding between CS, PEO and AC [124].

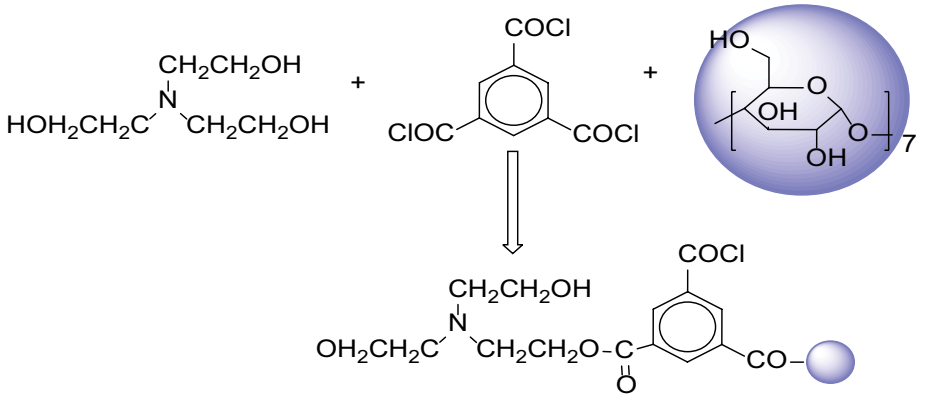
of a global water deficit. Membrane technology is increasingly being used to address municipal and industrial water supply and treatment needs. In desalination process, energy consumption and cost are the major elements for reducing the harmful environmental impact of desalinated fresh water supply, particularly in rural places with only a few options. The degree to which the role of membrane technology extends is primarily determined by its cost-effectiveness. Aside from desalination efficiency, concentrated discharge choices, energy consumption, health and environmental concerns are all major factors influencing the technology's adoption.

To satisfy the growing needs of the desalination industry, the membrane research community is focusing on constructing and developing a sustainable membrane that is ideally suited for desalination and can function in a variety of situations. While nanotechnology is maturing, new nanomaterials are actively engaged in the water purification field by removing impurities quickly and effectively. The coming together of membrane and nanomaterial research will almost certainly result in a new era of active membranes with a variety of interesting properties. A membrane with all the desirable characteristics to accomplish total rejection of waste and improve flux by multiple orders of magnitude would be an appealing proposition to industry.

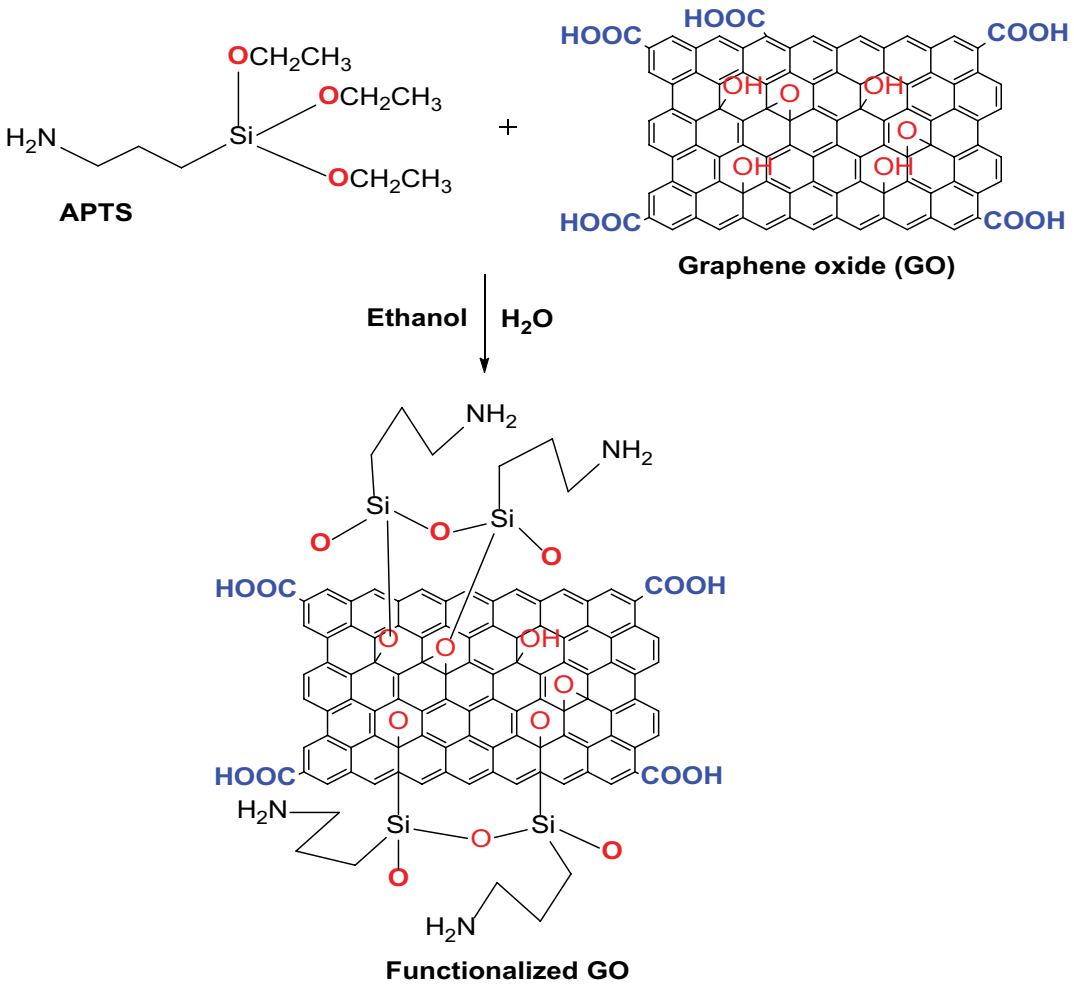
From an industrial viewpoint, resolving the challenges of a sustainable water future provide tremendous development potential as well as economic opportunities in water-related sectors. Despite the fact that the benefits of investing in these new membranes and membrane systems may not be immediately obvious, the loss of proficiency and reliability of traditional desalination technologies appears to exceed the investment cost. However, the cost-effectiveness of emerging technologies is a major factor in their acceptance. As a result, it is critical to integrate the technologies with as little disruption as possible to the current infrastructure. Retrofitting and modernizing existing desalination plants appears to become a more promising technique in this scenario. Incorporating innovative and sustainable materials and methods into existing facilities appears to be a cost-effective way to address tough water needs at a fraction of the expense of extending or constructing more costly water treatment plants. Developing the viability and flexibility of technologies has resulted in significant advancements in both the industrial and social worlds. Foremost membrane technology has progressed to the point where developing separation systems can provide high-quality fresh water with extreme reliability using more productive and cost-effective ways.

7. Conclusion

The growing freshwater demands require us to upgrade the technology with suitable modifications by blending polymeric membranes with different additives, to achieve high filtration performance for desalination and wastewater treatment. The performance of any polymeric membrane filtration purely rests upon physical characteristics of the additive blended with the membrane and leads to an increased number of functional groups inside and outside the surface of the membrane. Addition of polymers, metal oxides, and biopolymers depends upon the type of waste that needs to be eliminated from the water. The fabrication procedures are improving physical characteristics producing high quality membrane with cost-effectiveness. The commercial desalination treatments have adopted TFC and cellulose-based membranes for all bulk process. Ceramic membranes are also being used along with carbon-based membranes and metal oxide membranes with tunable surface area, porosity, and distribution effectively. This review sets a benchmark how the use of inorganic and organic blends in MMMs has led to their increased stability and wide scale application for seawater treatment. Despite all limitations, the use of nano-sized materials with polymeric membranes is significant for RO and NF processes for desalination and wastewater treatment which can potentially satisfy the growing water demands in future.



Scheme 12. Interfacial polymerization of TEOA and TMC with the β-CD [145].



Scheme 13. APTS and GO reaction pathway [161].

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