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Applications of nanotechnology in membrane distillation: a review study

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ABSTRACT

Membrane distillation (MD) is an effective water treatment process with relatively low cost compared to conventional membrane processes. This study investigates several factors affecting the performance of the MD membrane such as fouling, porosity, pore size, mechanical stability, contact angle, salt rejection, and other physical and thermal properties. Membrane performance can be improved if membranes are manufactured using nanotechnology. This work presents a review of the application of recently discovered nanotechnology that improves the properties and enhances the performance of membranes used in water distillation processes. The use of carbon nanotechnology-based membranes, nanoparticles, metal, and metal oxide nanocomposite is presented and discussed. The use of nanotechnology helps in making membranes less susceptible to fouling and compaction which results in more permeate flux. This study describes the use of scanning electron microscopy as a membrane characterization method and discusses the performance of MD under different operating conditions for the fabricated membranes by using nanotechnology applications. Due to the need for continuous improvement in membrane processes for water distillation and water treatment, the optimization of membrane performance and the parameters affecting this performance should be investigated.

Keywords: Membrane distillation; Nanotechnology; Permeate flux

1. Introduction

Most industrial membrane applications are water distillation, chemical treatment, and separation processes. In general, membrane separation processes can be divided with respect to the pressure gradient across the membrane into four categories: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis/forward osmosis (FO/RO). Membrane parameters with ranges of separation and with Membrane distillation (MD) operation principles are shown in Table 1.

As well as the operating conditions of the process, a membrane's physical and mechanical properties are important in determining the causes of membrane compaction and fouling. MD is widely used for water treatment [1–10] and separation processes [11]. Membrane separation processes

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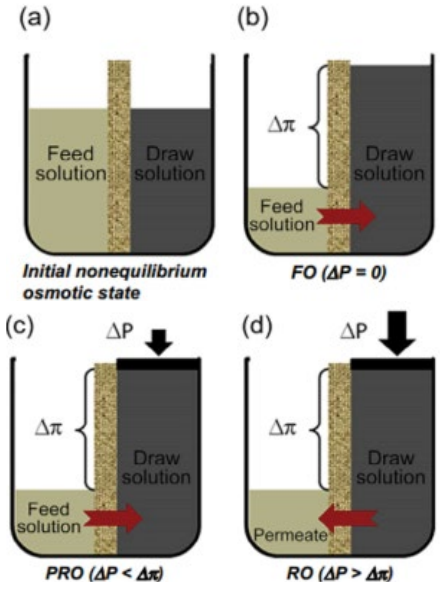
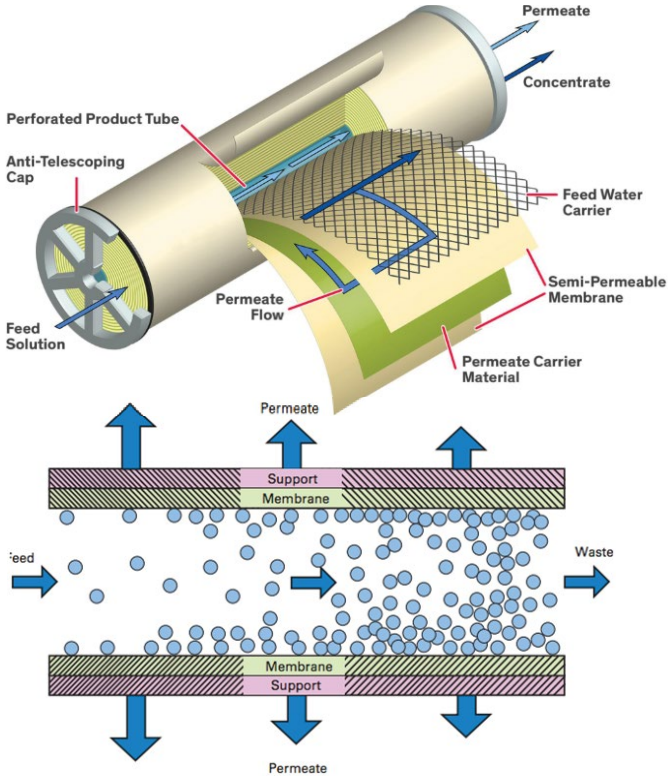
Table 1
Membrane features with ranges of separation [15] and MD operation principles [16]

Types	Reverse osmosis	Nanofiltration	Ultrafiltration	Microfiltration
Membrane	Asymmetrical	Asymmetrical	Asymmetrical	Asymmetrical
Thickness surface film	150–1 μm	150–1 μm	150–250 μm	10–150 μm
Pore size	<0.002 μm	<0.002 μm	0.02–0.2 μm	0.2–5 μm
Rejects	HMWC, LMWC, sodium, chloride, glucose, amino acids, proteins	HMWC, mono, di and oligosaccharides, polyvalent anions	Macromolecules, proteins, polysaccharides, viruses	Particulates, clay, bacteria
Membrane material(s)	CA – thin film	CA – thin film	Ceramic, PSU, CA, PVDF, thin film	Ceramic, PP, PSU, PVDF
Membrane module	Tubular, spiral – wound,	Tubular, spiral, wound	Tubular, hollow, fiber, spiral, wound, plate, and frame	Tubular, hollow, fiber, plate, and frame
Pressure	15–150 bars	5–35 bars	1–10 bar	2 bars<
MD operation principles [16]				

(a) Initial non-equilibrium osmotic state;
(b) Forward osmosis (FO) process (no pressure is applied);
(c) PRO process (application of hydrostatic pressure lower than the transmembrane osmotic pressure on the draw solution);
(d) RO process used for desalination of a saline feed solution (application of a hydrostatic pressure greater than the transmembrane osmotic pressure)

CA – cellulose acetate; PSU – polysulfone; PVDF – polyvinylidene fluoride; PP – polypropylene; HMWC – high molecular weight compounds: 100,000–1 million mol g^{-1}; LMWC – low molecular-weight compounds: 1,000–100,000 mol g^{-1}; macromolecules: 1 million mol g^{-1}.

are preferred among other technologies as they are inexpensive, easy to scale-up, and have low energy demand [12]. They can also be used to separate proteins, viruses, and gasses. Membranes play a part in the separation of organic chemicals. Such separation is known as organic solvent nanofiltration (OSN) membrane technology [13,14].

In the MD process, in order to reduce flux, the membrane should be hydrophobic [17] to avoid plugging its pores due to wetting (pore wetting). Thus, a hydrophobic membrane with high wetting resistance is required for better MD performance. Fouling is one of the most common problems which happens in MD after periodic use and affects membrane efficiency [18,19].

Current improvements to MD processes include the creation of new construction materials with better characteristics. The use of nanomaterials, either in membrane manufacturing or the direct use of nanoparticles in the fluid, results in better mass transfer processes in the membrane [20]. This review focuses on the use of different types of nanoporous membranes for water distillation with the aim of enhancing the performance of the membranes.

2. Review of nanoparticle use in the membrane fabrication

2.1. Importance of nanomaterial in membrane desalination

Traditional membranes suffer from many problems in desalination processes such as permeability, selectivity, chemical stability, and fouling. Such problems affect general performance in desalination. One particular graphene-based nanomaterial has a unique structure and has tunable physicochemical, biological, electrical, and mechanical properties that improve the performance of desalination processes that include desalination of water with high salinity (Na^+ and Cl^-). Carbon nanotubes (CNTs) have the same advantages in terms of excellent separation while graphene-based membranes are much easier to scale up with low-cost.

With regard to cost, producing distilled water by traditional desalination is currently more expensive. For example, the unit cost of RO seawater desalination is on average about US$2.0 m^{-3} compared to US$0.83 m^{-3} for desalination by nano-filtration. The high cost increases again when the price of energy goes up. Additionally, the high salt concentration in seawater imposes a thermodynamic limit of 1.1 kWh m^{-3}, and the theoretical minimum energy consumption at 50% recovery, significantly contributing to the overall cost of seawater desalination. Therefore, the development of high-performance desalination membranes using nanomaterials plays a key role in improving and developing membrane-based desalination technology. In fact, future membranes coupled with nanotechnology should have the following properties in order to be efficient for drinking water production: fouling resistance, low cost, high salt rejection, high water flux, and good mechanical stability [21]. Table 2 shows different applications of nanoparticles in membrane separation processes along with their costs.

Table 2
Cost of different application of nanoparticles in membrane water treatment

Membrane process	Membrane material	Wastewater type	Used nanoparticles	Cost
Ultrafiltration	Phosphatidic acid	Salt removal [22,23]	Zinc oxide (ZnO)	Cost for this type of project can typically run about 10%–15% of the cost of the entire project [25]
Ultrafiltration	Polysulfone	Bacterial removal from aqueous solutions [24]	Zinc oxide (ZnO)	
Microfiltration	Polyvinylidene fluoride	Heavy metal ions removal from wastewater [26]	Zinc oxide (ZnO)	Typical installation costs for microfiltration with a volume of 25 m^3 d^{-1}, amount to between € 25.000 and 50.000 For MF, one should assume an average operating cost of 0.1 to 0.15 € m^{-3} produced permeate [27]
Ultrafiltration	Polysulfone	Wastewater treatment, mainly for oily water [28]	Graphene	Reduction in energy consumption of 15%–46% which is reflected in the reduction of the high cost of energy, that is, 50% of the total water desalination cost [29]
Nanofiltration	Nitrocellulose	Water treatment for drinking applications [30]	Carbone nanotubes	Cost is estimated to be €0.214 m^{-3} of distilled water [31]
Nanofiltration	Polyamide cellulose	Heavy metal removal from wastewater [32]	Zinc oxide (ZnO)	Cost is estimated to be US$ 0.83 m^{-3} of treated water [33]
Forward osmosis	Polyvinylidene fluoride	Desalination [34–36]	Zinc oxide (ZnO)	Unit energy predicted cost of US$ 0.16 kWh for a demonstrated 25 MW osmotic power plant [37]

2.2. Nanomaterial type

Previous studies have shown the relevance of fullerene use and its water-soluble derivatives to enhance the properties of pervaporation membrane transport [38,39] and a cross-linking agent [40–42]. The porous structure of the membrane helps to improve mass transfer. Pore size is closely linked to membrane performance. Pore size determines the ability to resist mass transfer inside the membrane structure where the fluid flow inside the pores is described by different models such as Poiseuille flow and Knudsen diffusion [42–45]. Fig. 1 shows three different methods for adding nanoparticles into the membrane to improve the flow characteristics inside the membrane.

Dmitrenko et al. [47] presented new dense-mixed matrix membranes that were built by using poly(2,6-dimethyl-1,4-phenylene oxide) (PPO) and chitosan. The tested membranes were developed by using low hydroxylated fullerenol $C_{60}(OH)_{12}$ as nanoparticles. They found that in the case of chitosan, fullerenol behaved like a cross-linker.

Using fullerenol as a nano-modifier acted to increase water transport and enhanced the membrane's dehydration in two different polymer matrices by improving the properties of the pervaporation transport. Using fullerenol for the PPO and chitosan membrane as a cross-linker leads to an increase in flux and membrane selectivity. Fig. 2 presents two micrographs of dense membranes based on PPO and PPO-fullerenol (2 wt.%) composite [47]. The pure PPO membrane has a rough structure and heterogeneity that increases when adding fullerenol into the polymer matrix.

Table 3 shows the transport properties of dense, thermally cross-linked (1,400°C) and supported membranes based on chitosan and chitosan-fullerenol composite. Industrial ultrafiltration membranes based on polysulfonamide (UPM) and polyacrylonitrile (PAN) were selected as porous supports. The selectivity of the supported UPM membrane dropped to 94.05 wt. % of the water in the permeate compared to the dense chitosan membrane which was 97.35 wt.%. The supported membrane on the PAN had good selectivity which is about 96.38 wt.% of the water in the permeate.

Thus, the PAN supported membrane was chosen for further investigation and for modification using nanoparticles. The selectivity increased (98.37 wt.% water in the permeate) with nearly an identical flux by introducing 1 wt. % fullerenol into the chitosan matrix of the PAN supported membrane so an effective nanocomposite supported membrane was obtained from water impurities for purifying tetrahydrofuran.

Kusworo et al. [48] studied the performance of nano-hybrid-cellulose acetate (CA) CA/TiO_2 membranes used in eugenol treatment. The stability of fabricated membranes for OSN was investigated. The scanning electron microscopy image depicted in Fig. 3 indicates an asymmetric structure of the membrane sub-layer. It shows different values of the permeate fluxes for different nanoparticle loading, and a proportional relationship between the nanoparticles in polymer blend and an increase obtained in permeability.

To estimate the pore size, the molecular weight cut-off (MWCO) of the fabricated membrane should be determined. A dead-end filtration cell was used to find the pore size by applying different molecular weights (400; 600; 1,000; 4,000; 6,000; and 10,000 g mol^{-1}) with different chemical solutions. A visual handheld refractometer (Atago) was used to analyze the permeate sample. The MWCO was obtained at 90% solute rejection. The pore size radius in terms of the MWCO can be expressed as [49]:

$$r_m(\text{cm}) = 16.73 \times 10^{-10} \times (\text{MWCO})^{0.557} \quad (1)$$

The porosity of the membrane is one of the most important parameters, which is determined by measuring the weight difference between the wetted and dried membranes. The membrane porosity is then calculated as follows [48]:

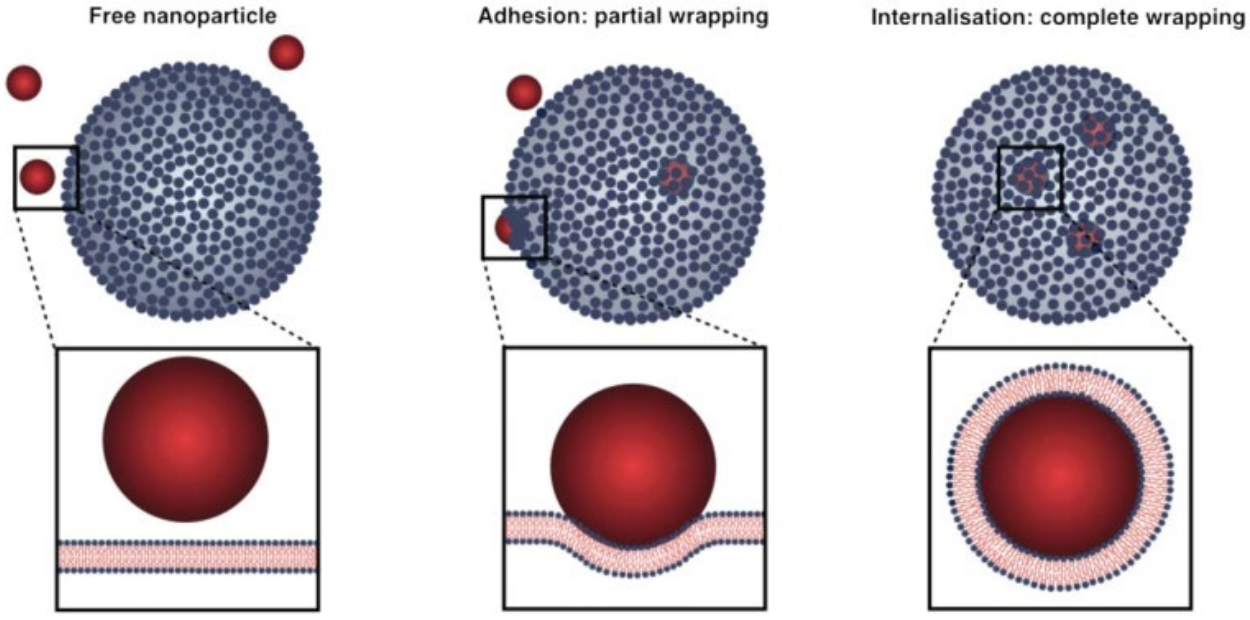


Fig. 1. Three possible NP adhesions to membranes [46].

Table 3
Dense and supported membranes used for pervaporation of mixture tetrahydrofuran (THF)-water azeotropic composition at 20°C based on chitosan and chitosan-fullerenol composite [47]

	Membranes (at 140°C for 100 min)	Flux (kg m^{-2} h^{-1})	Permeate, wt.%	
			Water	Ethanol
Dense	Chitosan	0.090	97.35	2.65
	Chitosan-fullerenol (1%)	0.063	99.34	0.66
Supported	Chitosan/UPM	0.133	94.05	5.95
	Chitosan/PAN	0.125	96.38	3.62
	Chitosan-fullerenol (1%)/PAN	0.099	98.37	1.63

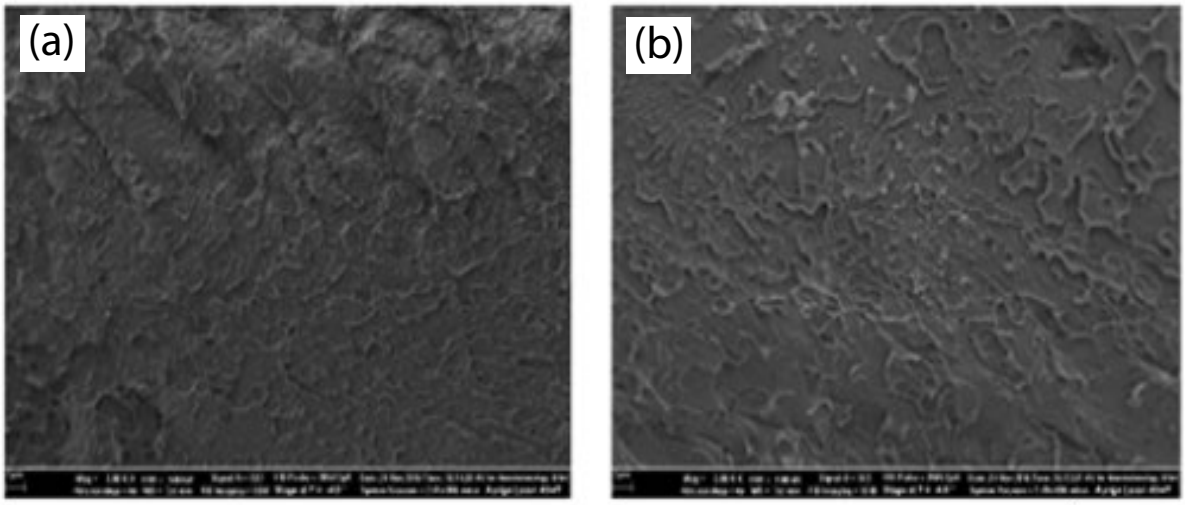


Fig. 2. Scanning electron microscopy (SEM) images of membrane cross-section based on (a) PPO and (b) PPO-fullerenol (2 wt.%) [47].

$$\varepsilon = \frac{w_{to} - w_{ti}}{\rho_w \times A \times l} \quad (2)$$

where ε is the membrane porosity, w_{to} and w_{ti} are the membrane weights before and after drying, respectively, A is the membrane surface area, l is the membrane thickness and ρ_w is the water density.

Fig. 4 shows the flux of clove oil for three membranes fabricated using nanoparticles. It shows that the increase in filtration time results in flux reduction due to membrane fouling. Initially, the flux of clove oil in the CA membrane was 23.4 L m^{-2} h^{-1} bar^{-1} before it sharply declines to 13 L m^{-2} h^{-1} bar^{-1} after 25 min of filtration. CA/nano-TiO_2 membranes yielded higher fluxes initially at 30 and 34 L m^{-2} h^{-1} bar^{-1} for nano-TiO_2 at 1 and 2 wt.% concentration, respectively.

Higher fluxes can be achieved because the nano-fillers prevent the formation of dead-end pores and retard compaction of the membranes, which makes CA/nano-TiO_2 membrane a favorable structure for separation processes. Moreover, the permeate flux was enhanced up to 52% by adding 2% nano-TiO_2 in the CA membrane compared to that of conventional ones.

Performance depends mainly on the membrane preparation conditions as well as on mechanical and thermal properties and the pore size and its distribution. The change in pore size distribution results in a significant change of membrane morphology and structure, and so a series of recent studies have reviewed membrane fabrication with different nanomaterials and techniques [50–57]. Other studies have focused on fabricating membranes by adding nanoparticles to improve membrane physical properties [58]. High permeability, high hydrophobicity, good mechanical properties, and low thermal conductivity are some of the important properties of MD membranes. Such properties are strongly affected by membrane preparation materials and procedures, as well as by the quantity and type of nanoparticles added.

These new, improved membrane types offer improved structure and functional properties for any separation processes. Organic-inorganic non-woven films are adapted to improve these properties. Nanoparticles as inorganic materials are added to the polymeric membrane matrix to improve the physical properties of the membrane. Metal oxides such as Al_2O_3, SiO_2, and TiO_2 may be used to enhance the physical properties [40] and silica-based porous mats can also be used [59,60] to improve physical properties.

Electrospinning is a simple method (Fig. 5) of preparing such materials. Here nanometric fibers can be fabricated for both composite and polymeric materials [61–67].

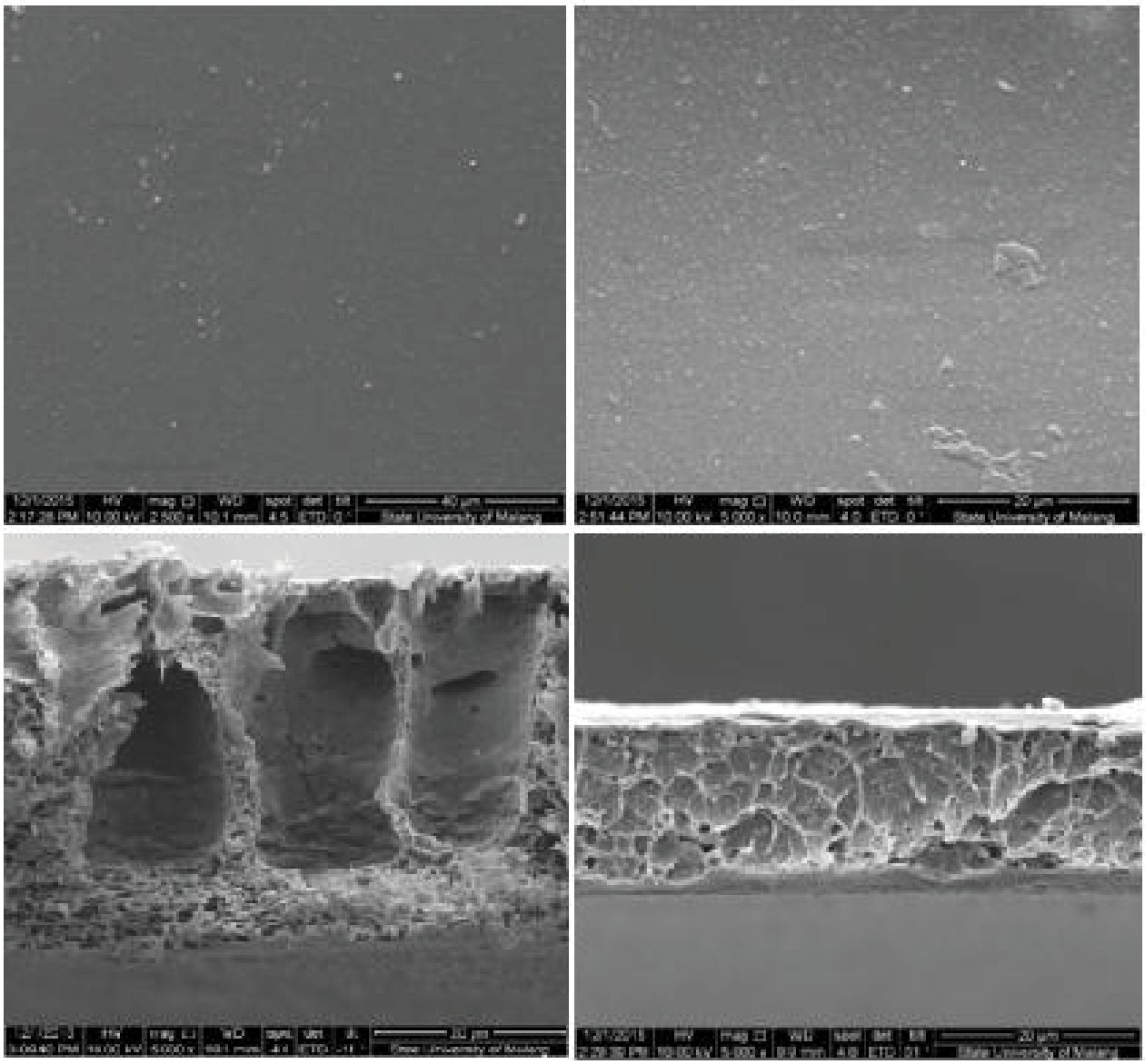


Fig. 3. SEM image of fabricated membrane used in eugenol treatment [48].

When nanoparticles were used, the applied procedure has led to the fabrication of different membranes. Many physical properties are characterized for every single membrane such as porosity, mean pore size, young modulus, liquid entry pressure (LEP which is defined as the pressure at which the first drop of the feed solution appears on the permeate side), and contact angle. These properties were measured under different concentrations of tetraisopropyl orthotitanate (TTIP). The physical properties are shown in Table 4.

By changing the spinning conditions, different electrospun fibers can be developed with different pore sizes and thicknesses of porosity for liquid separation [68]. Polyvinylidene fluoride (PVDF) nanofiber was electro-spun into the membrane and desalinated with NaCl. The salt rejection rate was 99.9% [69]. It should be noted here that the morphology of the fibers and their thickness can be changed by changing the spinning time because this will increase the surface roughness of the PVDF membrane with a high contact angle of 130°. By proper spinning, the nanoparticles can be embedded within the fiber [70–72]. Electro-spin web was used for the growth of biological cells which would result in great membrane potential for nanofiltration [73–75]. Moreover, clay nanoparticles have been reported in the electro-spun fiber [76]. The polytetrafluoroethylene (PTEE) membrane was exposed to N_2/H_2 plasma in the dose matrix, which resulted in improving the membrane permeation and reducing the contact angle and energy cost [77].

The porosity of the fabricated membranes can be obtained from:

$$\varepsilon = \frac{\left(w_{\text{to}} - w_{\text{ti}}\right)/\rho_1}{\left(w_{\text{to}} - w_{\text{ti}}\right)/\rho_1 + \left(w_{\text{ti}}\right)/\rho_p} \tag{3}$$

where w_{to} and w_{ti} are the weight of the wet and dry membranes, respectively, ρ_l is the isopentane density, and ρ_p is the polymer density.

Other work has been carried out using the fabricated membranes in water distillation to purify the water of heavy metals. The experiments were conducted at a permeate applied vacuum and constant feed flow rate. Fig. 6 presents the results of three different types of fabricated membranes.

Fig. 6 shows that the increase in feedwater temperature results in an increase in the permeate flux, this is due to the fact that the increase in temperature results in higher vapor pressure. Moreover, the figure also shows that permeate flux increases as the feed flow rate increases. There are two main reasons for this; first, the permeate velocity increases, which in turn increases Reynold's number resulting in better convective heat transfer performance, and second, the resistance of the boundary layer decreases as the permeate velocity increases.

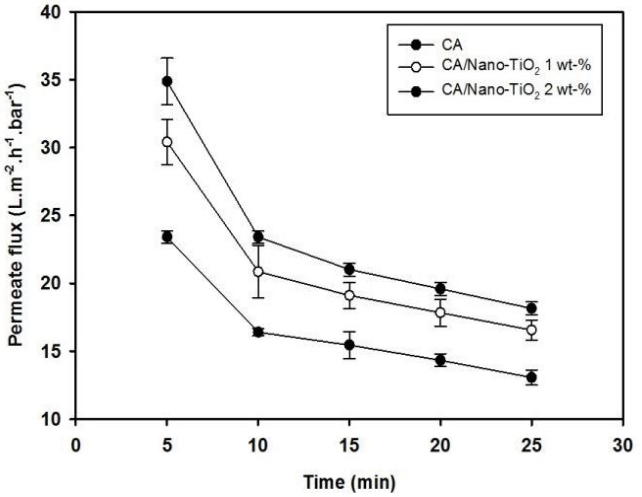


Fig. 4. Clove leaf oil flux profiles permeate for each membrane [48].

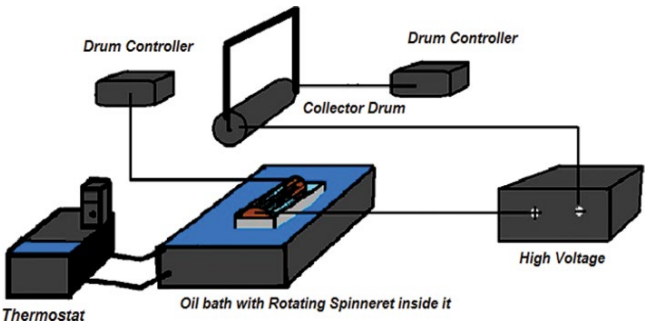


Fig. 5. Needle-less electrospinning setup schematic used to produce nanofiber membranes [58].

The performance of the membrane was enhanced by the application of the Ti–O–Ti structure into the PVDF electrospun membrane in terms of durability and other physical properties. However, it did not improve the permeate flux of the membrane. However, by using asymmetric thermal treatment of hybrid membrane, an enhancement in the membrane permeate flux was obtained without affecting the concentration and separation performance.

The nanoparticles added to the membrane could not only reduce the fouling but also make the membrane resistant to fouling. It was also referred to in the literature that nanoparticles enhanced the membrane performance by improving the thermal, mechanical and chemical stability, which would result in increasing the salt rejection and increasing the permeate flux [78–83].

3. Different types of nanoporous membranes for MD

3.1. Inorganic-based membranes

3.1.1. Ceramic membranes

Ceramic membranes are artificial membranes made from inorganic materials such as titanium, alumina, zirconium, iron oxides, silicon carbides, or some glassy materials [84–89]. They are used for liquid filtration [90]. In contrast to polymer membranes, they are more homogeneous (Figs. 8a and b). Aluminum polysilicates and natural clay are extruded to form a porous tubular thin membrane [91]. The advantage of ceramic membrane over the organic membrane is because of its high mechanical and thermal stability over a wide pH range. It is applicable for filtration with a wide range of solvents at high temperatures [92]. Ceramic membranes are tubular with a large surface and they are hydrophilic in nature to make them operating easily with the hydrophobic group [93,94]. By sol–gel method, alkoxysilane or fluoropolymer membranes are prepared by grafting where the hydroxyl group presented on metal oxide is used for the reaction with a functional group of oligomers [91,95–97]. Ceramic materials such as zeolite and silica are able to provide the required properties for desalination though they are partially tolerant of water [98–101]. Table 5 presents the performance of inorganic membranes for different membrane geometries.

Table 4
Comparative analysis results for various prepared membranes [58]

Membrane	TTIP conc. (wt.%)	LEP (kPa)	CA (°)	Porosity (%)	Mean pore size (nm)	Young modulus (MPa)
M1	0	75 ± 4	124 ± 2	73 ± 3	300 ± 15	400 ± 50
M2	5	110 ± 5	125 ± 2	72 ± 3	325 ± 10	450 ± 70
M3	10	125 ± 5	130 ± 3	74 ± 3	320 ± 10	490 ± 50
M4	20	140 ± 4	135 ± 3	75 ± 3	270 ± 15	550 ± 40
M3 (treated)	10	155 ± 5	145 ± 5	72 ± 3	320 ± 10	510 ± 40

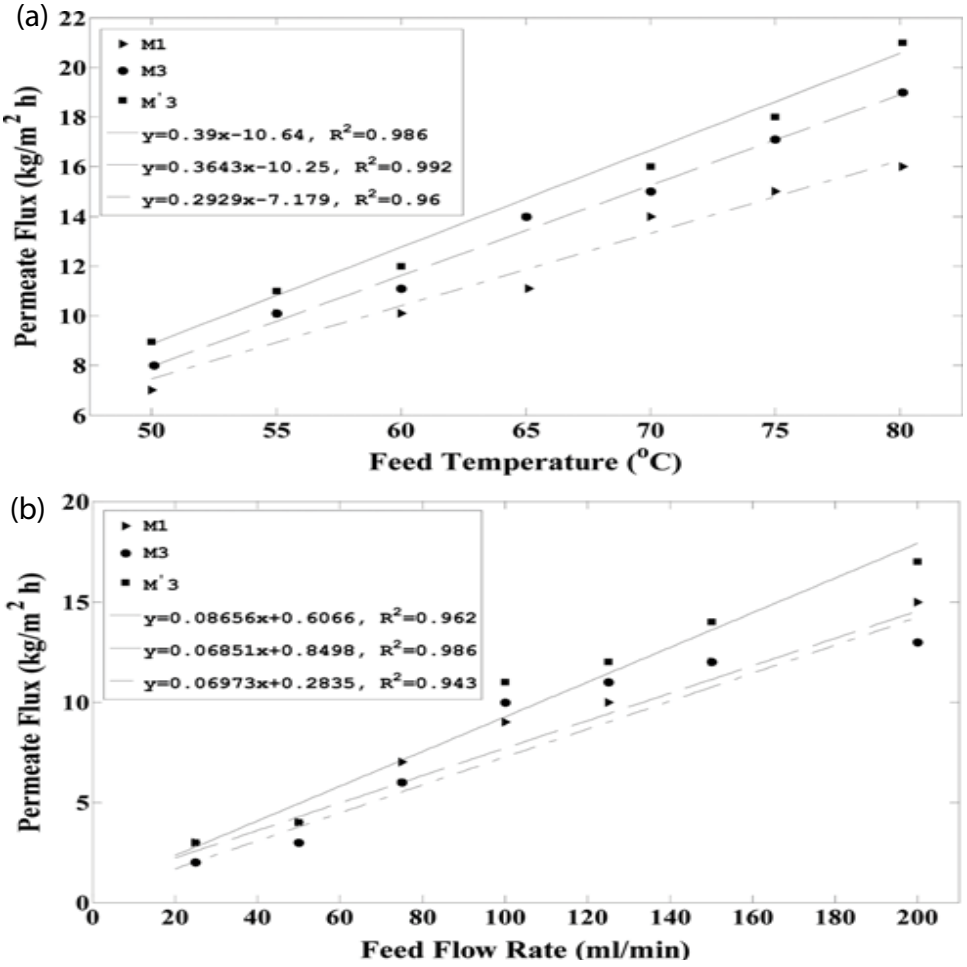


Fig. 6. (a) Feed temperature and (b) feed flow rate effects on permeate flux [58].

Table 5
Performance of inorganic membranes [102]

MD configuration	Material	Geometry	Maximum flux (kg $m^{-2} h^{-1}$)	Driving force (kPa)	Reference
AGMD	Alumina-fluorosilane functionalized	Tubular	6.02–6.76	70	[96]
DCMD	Alumina-silanized	Flat disc	7.8–8.1	12.23	[97]
VMD	Titania (5)	Tubular	6.08	0.3	[92]
VMD	Zirconia (50)	Tubular	7.5	0.3	[93]
AGMD	Zirconia (50)	Tubular	2.7–4.7	38.5–83.9	[93]
DCMD	Zirconia (50)	Tubular	1.7–3.95	38.5–83.9	[93]
AGMD	Alumina	Tubular	5.39	70	[97]
AGMD	Zirconia	Tubular	2.8–6.9	70	[97]
AGMD	Alumino-silicate	Tubular	5.08	83.9	[94]
AGMD	Alumina	Tubular	4.91–5.04	83.9	[94]
AGMD	Zirconia clay with perfluorodecyltriethoxysilane	Tubular	5.08	83.9	[94]
AGMD	(Pore size 15 nm) clay with perfluorodecytriethoxysilane	Flat disc	3.95–5.83	47.36	[91]
AGMD	(Pore size 180 nm)	Flat disc	5–7.2	47.36	[91]
VMD+	Alumina	Flat sheet	0.72	47.36	[98]
VMD+	Silica	Flat sheet	1.7		[98]

3.1.2. Carbon nanotube membranes

Metal and metal oxide nanocomposite membranes with nanomaterials are used to improve the MD process where many scientific studies are available in the literature [103–106]. Carbon nanotechnology-based membranes are types of membranes where nanoparticles are involved in the preparation of the membrane materials. CNT is a nanomaterial and an allotrope of carbon which is used due to its excellent properties such as surface adsorption, low energy consumption, high selectivity, and water flux, chemical, mechanical, and thermal properties [107–110]. Graphene is a good example of CNT [111–113]. Graphene is known to be hydrophobic material and possesses a high surface area compared to its weight, thus providing the desired mechanical and thermal properties for making the membrane stable during thermal and mechanical fluctuating conditions. Rikhtehgaran and Lohrasebi [114] investigated the performance of using multilayer nanoporous graphene (NPG) membranes for water purification with different layers separation, pore size, and number of layers. Their results showed that water flux through the membrane and salt rejection were significantly dependent on the pore size and number of layers of graphene.

NPG membranes are considered in producing freshwater by using RO desalination. They can make the desalination process more economical with better filtration efficiency [115,116]. Graphene as a carbon nanostructure is also used in designing carbon-based devices [117–119] and it has been studied experimentally [120–122] and computationally [7,123–126] for saline water filtration. Graphene can be employed for a high rate of salt rejection, a high rate of water transport in desalination [127–132], and a reduction in membrane thickness [5,130] that leads to lower pressure drop and higher filtration efficiency. Compared to zeolite-based membranes, graphene has higher flux rates [7,82,112,123,129,130,133]. Graphene oxide (GO) is the hydrophilic oxidized form of graphene [134–136]. Preparation of GO is reported in the literature which shows that GO has been widely used in different applications [137–139]. Hummers [137] and Staudenmaier [140] have improved the preparation method of GO which was firstly prepared by Brodie [141]. The applications of CNT in fabricated nanocomposite membranes are summarized in Table 6.

The inclusion of nanotechnology in developing membrane leads to nanoparticles (NPs) composite membranes. As mentioned above, the functionalized GO/PVDF membrane [140] lies under this type. Hydrophilicity was found to be improved by increasing NP's content which was checked by contact angle measurement [162]. The TiO_2–SiO_2/PA/PSf (PSf – polysulfone and PA – polyamide membrane) nanocomposite membranes were prepared by polymerizing the PA layer onto the supported PSf membrane and allowing its reaction with 3-aminopropyltrimethoxysilane. Then they were dipped into TiO_2 dispersed in the organic phase. Fig. 7 illustrates the main mechanism of modifying the membrane surface.

The hydrophilicity effect on the bovine serum albumin rejection of PVDF/GO and PVDF/MWCNT (multi-walled carbon nanotubes) nanocomposite membranes are shown in Fig. 8 [156].

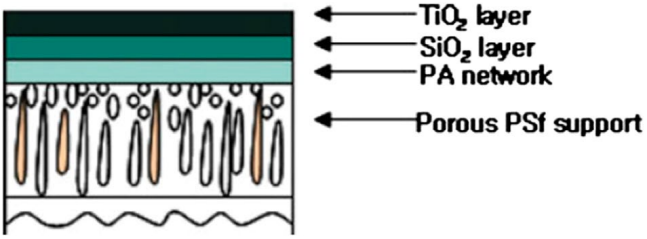


Fig. 7. Schematic of PSf membrane surface modification with nanoparticles composite and polyamide [142].

Table 6
Membrane modification using carbon nanotechnology-based NPs [142]

Membrane base material	Material/method	Reference
PSf modified	MWCNTs/blending	[143,144]
BPPO	MWCNTs/blending	[145]
PP and PES	MWCNTs/surface coating	[146]
PES	MWCNTs/blending	[147]
PAN	MWCNTs/blending	[148]
Chitosan (CS) coated PES	MWCNTs/blending with CS	[149]
PVDF	GO/blending	[150,151]
PSf	GO/blending	[152–154]
PVDF	GO-oxidized MWCNTs (OMWCNT)/blending	[155]
PVDF	GO-MWCNT/blending	[156]
PES	GO/blending	[157]
PVDF	Functionalized GO/blending	[158]
PVDF	GO nanoplatelets/blending	[159]
PVDF	GO/water bath coagulation	[160]
PES	Functionalized MWCNTs/blending	[161]

BPPO is brominated polyphenylene oxide, PVDF is polyvinylidene fluoride, PES is polyethersulfone and PSf is polysulfone.

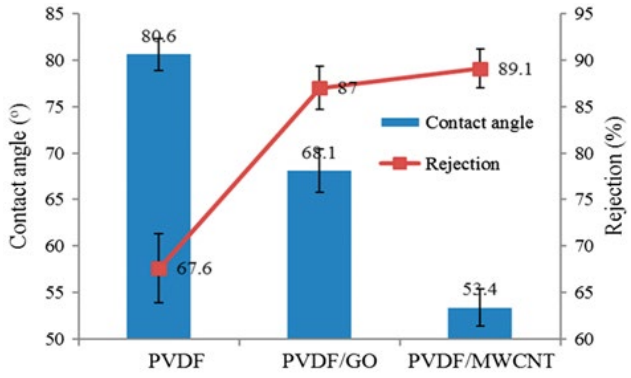


Fig. 8. Contact angle and rejection values plotted for different membranes (data used from [156]).

During the last decades, there has been growing interest in the development of different types of membranes composed of CNT materials including bucky paper (BP) and composite material form for a wide range of filtration applications. They have extraordinary thermal conductivity, mechanical and electrical properties [163,164]. The BP has a paper-like structure in which CNTs are held together by Van der Waals forces. The properties and performance of the CNT's membranes for water desalination are given in Table 7 [102].

Multilayer BP membranes were produced with chemical vapor deposition grew carbon tubes and coated with polystyrene and PVDF to enhance their mechanical stability over time, without drastically changing their average pore size and porosity [164]. CNTs were also modified by grafting with alkoxysilane in order to enhance the hydrophobicity of their surface area [165]. BP surface was coated with PTEE in order to lower the membrane surface energy and increase the hydrophobicity and life span [166].

3.2. Organic-based membrane

3.2.1. Polymeric membrane

Organic membranes are mainly formed of polymeric materials as polyimide, polyvinyl alcohol, polyethersulfone (PES), polypropylene (PP), CA, PVDF, cellulose nitrates, PAN, polytetrafluoroethylene (PTFE), polysulfone (PSU) and biomacromolecules [167–174]. Hydrophobic PVDF-PTEE, PES, CA, and PE hollow fiber membranes for desalination were successfully fabricated and commercialized by various companies as shown in Table 8 [102,175]. The hydrophobicity of PVDF was very high, which was enhanced by increasing the fluoride ratio [176]. The CA membranes were used as support for the deposition of hydrophobic materials [177].

3.2.2. Hydrophilic/hydrophobic membrane in direct contact membrane distillation

For the first time in the 1990s, co-extrusion was applied for the fabrication of dual-layer hydrophilic/hydrophobic hollow fiber, especially for direct contact membrane distillation (DCMD) process. Hydrophilic and hydrophobic particles were incorporated into the outer and inner layer dope solutions, respectively in order to enhance the heat polarization effect which was stronger than that of the MD process [177]. The wetting on the permeate side reduced the temperature polarization and helped in the diffusion of heat and condensation of water vapor into the bulk permeate water [18,180–185]. Dual-layer membranes were mostly processed as hollow fiber (HF) [18,182]. The hydrophobic ultrafiltration PES membrane was modified by CF4 plasma surface. This modification changed the hydrophilic membrane into hydrophobic for MD [186]. The plasma modification converted the contact angle of the hydrophilic membrane from 0° to 120°. A stable membrane was then obtained with no leakage and with high water flux. The PVDF membrane was coated with TiO_2 followed by fluoro-silanization of the surface to increase the multi-level roughness and reduce the surface free energy [187]. The membrane was changed from a common hydrophobic state to a superhydrophobic state.

3.3. Hybrid and exotic membranes

3.3.1. Mixed matrix nano-composite membrane

Adding inorganic materials into a polymeric matrix would result in producing hybrid membranes [188–194]. A PVDF-HF surface was grafted with graphene particles multi-layer CNT [181] to enhance the thermal conductivity. The nano and macroparticles reduced the heat diffusion of the membrane. An increase in the surface roughness of the membrane affected the surface heat conditions and the contact angle. Fig. 9e shows the results of the mathematical modeling investigating the effects of thermal conductivity on vapor flux.

Table 7
Properties and performance of the CNTs membranes for water desalination [102]

Sample	Porosity thickness		Pore size (nm)	Contact angle (°)	Flux (kg h^{-1} m^{-2})	Salt rejection (%)	Dp (kPa)	Permeability (×10^{-8} kg m^{-1} h^{-1} Pa^{-1})
	(%)	(μm)						
Self-supporting BP	90	55	25	118	12	94	40.43	1.63
Sandwiched BP	90	140	25	105	15	95.5	55	3.81
PTFE coated BP	88	105	25	155	7.75	99	78	1.04
Alkoxy silane functionalized BP	90	62	23	140	9.5	98.3	35	1.68

Many experiments have been done to alter the surface energy of polymer membranes. Several of these resulted in drastic changes in surface roughness. Increases in hydrophobicity and decreases in thermal conductivity of membrane surface were due to the reduction in contact angle between the membranes and the liquid streams [180,181].

CNT's immobilized membranes were developed worldwide during the last decades. The CNT increases the permeability of substance through the membrane and enhances its selectivity. CNT has a high thermal conductivity which results in reducing the temperature gradient of the membrane pore and enhances the performance of MD [195].

4. Conclusions

This paper presented a review of nanotechnology applications in MD processes. The review showed that whatever the efficiency value of the MD, it would suffer from traditional problems such as membrane compaction and fouling. The study showed that nanotechnology provided solutions, improving and upgrading the performance of MDs. These improvements included providing better mechanical and physical properties and enhancing MD performance (such as allowing higher permeate flux). The application of nanotechnology in MD can include the use of metal oxide NPs in hydrophobic membranes to reduce fouling.

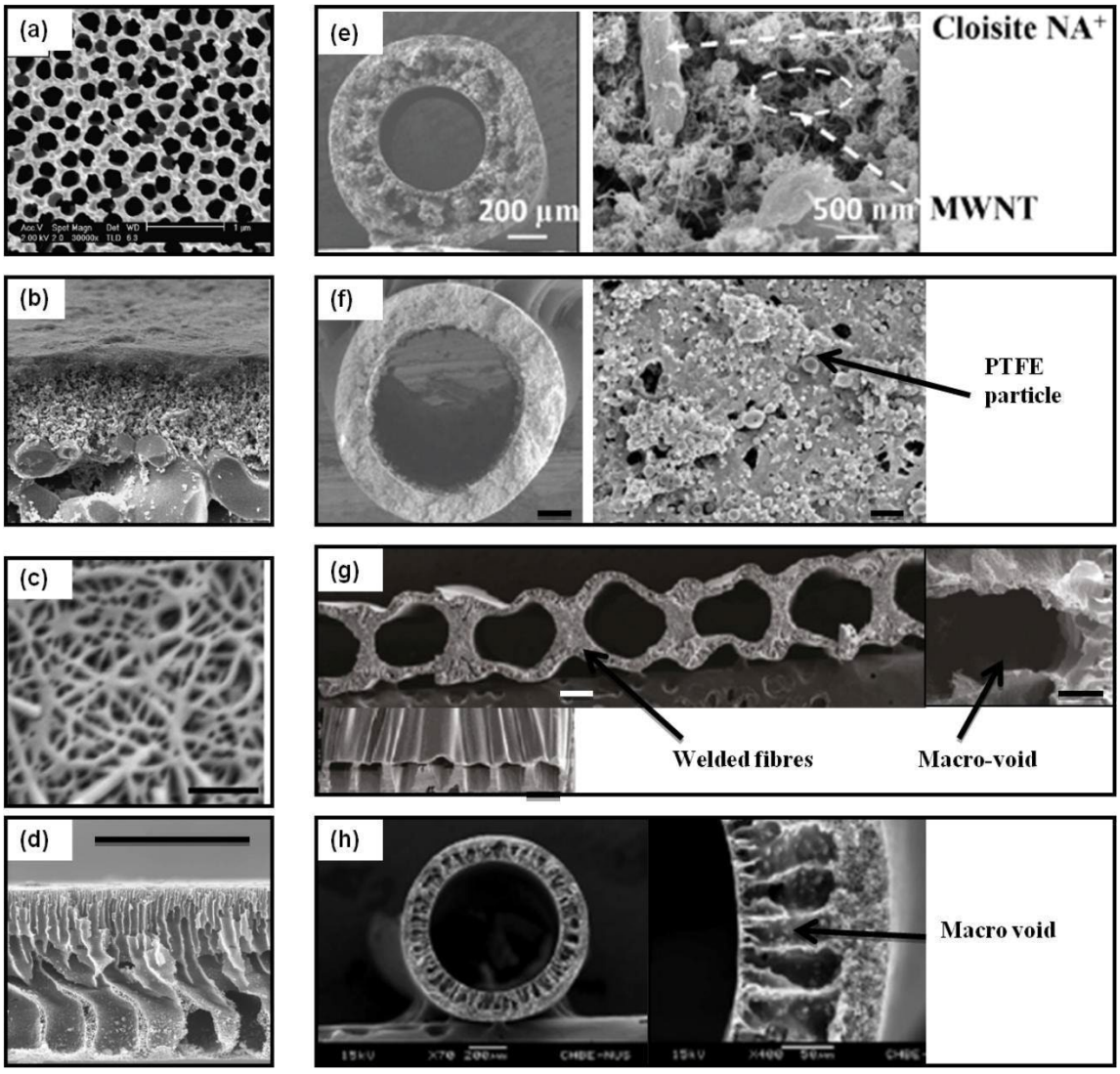


Fig. 9. Morphology of membranes used in MD [102] flat sheet (left column) and hollow fibers (right column). (a) Hendren et al. [95] anodisc polymeric ferric sulfate surface treatment, (b) Krajewski et al. [96] zirconia supported alumina membrane, (c) Dumée et al. [164] PTFE coated carbon nanotube bucky paper—scale bar corresponds to 400 nm, (d) Qtaishat et al. [179] used polysulfone as a base material and modified the surface with different amounts of fluorinated macromolecules (M4 membrane—scale bar corresponds to 100 µm), (e) Su et al. [182] added graphite particles and carbon nanotube to a PVDF/PAN blend (M3 membrane—overall shape and inner layer), (f) Teoh et al. [195]—PVDF/PTFE composites 50 wt. PTFE particles—overall HF view and inner layer—scale bar is 100 µm (left) and 1 µm right, (g) Teoh et al. [195] grooved membranes—PVDF multichannel membranes—scale bars: top 300 µm and bottom 500 µm, and right 5 µm, and (h) Wang et al. [185]—super high flux membranes D3—scale bar left 200 µm right 5 µm.

Table 8
Examples of commercial membranes used in MD [102]

Product	Manufacturer	Material	Support	Pore size (μm)	LEP (kPa)	Reference
TF200	Gelman/Pall (New York, USA)	PTFE	PP	0.2	282	[3]
TF450	Gelman/Pall (New York, USA)	PTFE	PP	0.45	138	[3]
TF1000	Gelman/Pall (New York, USA)	PTFE	PP	1	48	[3]
	Pall (New York, USA)	PTFE	PET	0.02	1585	
Emflon	Pall (New York, USA)	PTFE	PET	0.2	551	[178]
	Pall (New York, USA)	PTFE	PET	0.45	206	
	Pall (New York, USA)	PTFE	PET	1	137	
FGLP	Millipore (Massachusetts, USA)	PTFE	PE	0.2	280	[3]
FHLP	Millipore (Massachusetts, USA)	PTFE	PE	0.5	124	[3]
	Gore	PTFE	PP	0.2	368	
Gore filtration media	Gore	PTFE	PP	0.45	288	[3]
	Gore	PTFE	PP	0.2	463	
GVHP	Millipore (Massachusetts, USA)	PVDF	None	0.22	204	[3]
	Millipore (Massachusetts, USA)	PVDF	None	0.45	105	
	Membrane solutions	PTFE	PP	1.0	24	
HVHP	GE	PTFE	PP	0.22	154	[3]
	GE	PTFE	PP	0.45	91	
	GE	PTFE	PP	1.0	48	

PP, PE, PET respectively correspond to polypropylene, polyethylene, and polyester. The contact angle on PVDF and PTFE was reported to be 90° and 140° for surface energies of 30.3 mN m^{-1} and 9–20 mM m^{-1}, respectively at 20°C [179].

TiO_2 NPs could be used as composites with GO, MWCNT, SiO_2, and Ag. Carbon-based nanomaterials could be functionalized and hydrophilized with metal/metal-oxide NPs to improve the chemical properties of the latter while imparting the mechanical properties of the former. The addition of nanoparticles in membrane fabrication for use in many distillation applications such as MD, RO, MF, UF, and NF results in improved membrane performance. Further study of how to manufacture membranes using nanotechnology is recommended, especially of the type of nanoparticles used. Moreover, the operating conditions of the fabrication of nano-membranes should be clearly determined based on the type of desalination process used.

Symbols

r	—	Radius, m
w	—	Weight, kg
ρ	—	Density, kg m^{-3}
A	—	Area, m^2
l	—	Thickness, m

Greek

ε	—	Porosity

Subscripts

m	—	Membrane
to	—	Before drying
ti	—	After drying
w	—	Water
1	—	Isopentane
p	—	Polymer

Abbreviations

MWCO	—	Molecular weight cut-off

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