Advances in Membrane Materials and Processes for Desalination of Brackish Water

Summited to

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Abstract:

Purpose of Review This review aims to succinctly summarise recent advances of four key membrane processes (e.g. reverse osmosis (RO), forward osmosis (FO), electrodialysis (ED), and membrane distillation (MD)) in membrane materials and process designs, to elucidate the contributions of these advances to the steadfast growth of brackish water membrane desalination processes. With detailed analyses and discussions, the ultimate purpose of the review is to shed light on the future direction of brackish water desalination using membrane processes.

Recent Findings Brackish water has widely varying particulate matter and boron contents, posing great risks of membrane fouling and excessive boron levels to the membrane desalination processes. Recent advances in these four membrane processes largely focus on improving fouling resistance, boron rejection, water flux, and energy efficiency. Aquaporin membranes and thin-film composite polyamide membranes incorporated with nanoparticles exhibit excellent performances for RO and FO, whereas super-hydrophobic membranes prove their great potentials for MD. While recent advances in RO and ED process designs are orientated towards membrane fouling prevention by exploring respectively novel energy-saving membrane-based pre-treatment and reversal operation, recent studies on FO and MD are centred on reducing the energy costs by advancing the fertiliser-drawn concept and utilising waste heat.

Summary Membrane processes are dominating brackish water desalination, and this trend is hardly to change. Membranes based on nanoparticles and other novel materials are deemed the next membrane generation, and innovative membrane process designs have demonstrated great potentials for brackish water desalination. Nevertheless, further works are needed to scale up these novel membrane materials and designs.

Keywords: brackish water desalination; membrane processes; reverse osmosis (RO); forward osmosis (FO); electrodialysis (ED); membrane distillation (MD).

1. Introduction

Desalination has become a viable alternative fresh water supply in many water-scared areas worldwide [1-4]. Currently, large-scale brackish water and seawater desalination plants around the world provide 95 million m^3 of fresh water per day, meeting the daily demand of more than 1% of the global population [4]. Given recent technological advancements, desalination processes have become significantly more energy-efficient and cost-effective. For example, the invention of pressure recovery devices has markedly reduced the energy consumption and the operational cost of the reverse osmosis (RO) process, rendering brackish water and seawater RO desalination a technically and economically viable supply of fresh water [5-7]. Compared to seawater, brackish water offers a more cost-effective fresh water supply because of its considerably lower salinity and inland location. As a result, half of desalination plants worldwide rely on brackish water in spite of its distinctly limited availability compared to seawater [4].

The global brackish water desalination market is largely dominated by membrane processes [6, 8-10]. The membrane desalination processes do not require the phase change of water to achieve the salt-water separation. Instead, they deploy membrane to facilitate the removal of salt from water, thus desalting saline waters with significantly less energy compared to thermal distillation desalination. The membrane desalination processes are also more compact and have smaller physical footprints than the thermal distillation ones. In other words, the membrane processes offer more cost-effective and energy-efficient desalination means for fresh water provision, particularly from brackish water. As a result, most of brackish water desalination plants worldwide use membrane processes as their core technology [4].

This paper aims to provide a comprehensive review on recent advances in membrane processes and materials destined for brackish water desalination. The membrane processes reviewed in this paper include pressure-driven RO, osmotically driven forward osmosis (FO), electrically driven electrodialysis (ED), and thermally driven membrane distillation (MD). The review starts with an analysis of brackish water characteristics to highlight the advantages of and challenges to the membrane processes for brackish water desalination. The review then thoroughly discusses the recent advances in membrane materials and process designs orientated towards brackish water desalination of each membrane process. The ultimate purpose of the review paper is to shed light on the future directions of brackish water desalination using these membrane processes.

2. Characteristics of brackish water

Brackish water is defined as water with salinity in the range of 1,000–15,000 mg/L [11]. Given this salinity, brackish water needs to be reduced to fresh water (i.e. with salinity ≤500 mg/L) via a desalination process to be usable by humans and plants. The characteristics of brackish water, including salinity, temperature, and potential membrane foulant concentrations, strongly affect the selection and performance of the desalination process.

Brackish water salinity and temperature vary greatly with weather and geological location [11-14]. For example, the brackish water feed to the Gran Canaria desalination plant in Spain has salinity widely varying from 2,100 to 8,000 mg/L throughout the year [14]. Similarly, the salinity of brackish water feed to the desalination plant in Morocco changes from 650 to 1,300 mg/L during a year due to water evaporation and rainfall dilution [12]. The brackish water RO desalination plant in Morocco also suffers from seasonal feed water temperature change (i.e. 10–22 °C), leading to a 30% variation in the process water flux [12]. The variations in brackish water salinity and temperature have crucial implications for most membrane desalination processes because their performance indicators (e.g. water flux, salt rejection, energy consumption, and fouling propensity) are critically dependent on feed water salinity and temperature [11].

Brackish water is characterised as water sources with fluctuated particulate matter content [13, 15]. Unlike in seawater, particulate matter in brackish water is originated from natural and human-induced sources including erosion of stream bank and runoff from agricultural lands and production sites [15]. As a result, brackish water particulate matter content (i.e. turbidity) markedly differs depending on the season and geological location. For example, brackish water sourced from the Niger Delta, Nigeria has turbidity widely varying from 2.5 to 26 NTU [13]. The wide variation in brackish water turbidity poses a great risk of membrane fouling, and hence exerts strong influences on the design and operation of the brackish water membrane desalination processes.

Highly deviated boron concentration is another notable characteristic of brackish water [16-19]. While seawater has a stable boron concentration of around 4.6 mg/L, the boron concentration in brackish water wildly varies from 0.3 to 100 mg/L [17-19]. In brackish water, boron in the form of uncharged boric acid has a very small hydrated radius; therefore, it can penetrate through the membrane and contaminate the water product [20]. Indeed, commercial RO membranes are unable to completely remove boron from brackish water, and it has been well-recognised that boron contaminated water can lead to detrimental health and ecological

consequences [20]. Therefore, advances in membrane materials and process are critical for the brackish water RO desalination to meet the stringent regulations of boron level in desalted water.

3. Recent advances in membrane materials and processes for desalination of brackish water

3.1. Reverse osmosis (RO)

The pressure-driven reverse osmosis (RO) desalination process relies on a dense, semi-permeable membrane and a high hydrostatic pressure to achieve the salt-water separation. The dense RO membrane is selectively permeable to water while rejecting most dissolved salts and suspended solids. When the membrane separates brackish water and fresh water, under natural osmosis water migrates through the membrane to dilute the feed. To reverse the migration of water across the membrane, brackish water RO desalination applies a high hydrostatic pressure on the feed side (Fig. 1). The RO process water flux depends on membrane water permeability (A), the brackish water osmotic pressure, and the applied pressure as expressed below [6]:

$$J_{water} = A \times (\Delta p - \Delta \pi) \quad (1)$$

where Δp is the applied pressure and $\Delta \pi$ is the osmotic pressure difference between the brackish water feed and fresh water.

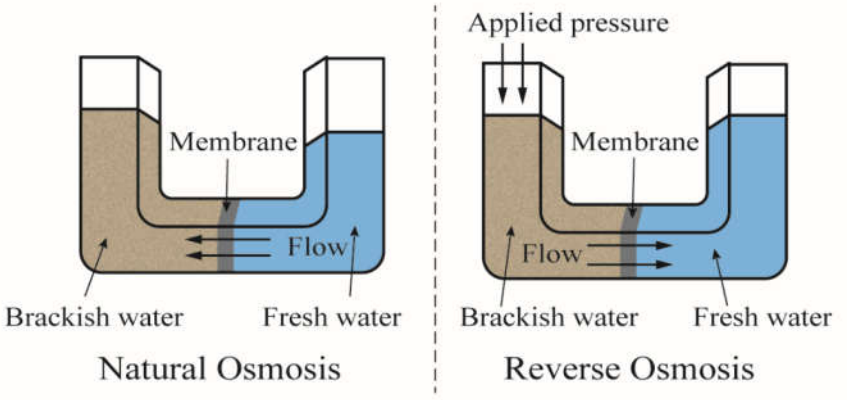

Fig. 1 The illustration of water migration in a natural osmosis and reverse osmosis process (adapted from [21]).

The efficiency of the brackish RO desalination process is reflected by the quality and cost of product water. Commercial RO membranes reject mostly all virus, bacteria, and divalent ions, while achieving above 96% rejection of monovalent salts. Therefore, RO desalination of brackish water effectively meets the regulations for fresh water supplies. However, the limited

142 removal of small-molecule contaminants such as boron remains a bottleneck for the practice of
143 brackish water RO desalination for drinking water [22].

144 The cost of brackish water RO desalination is composed of capital investment and
145 operation/maintenance costs, and strongly affected by the feed water salinity. Indeed, given its
146 low salinity, brackish water RO desalination offers a lower desalted water cost than seawater
147 RO desalination. For example, at the same capacity, the desalted water cost of brackish water
148 RO can be a half of that of seawater RO [23]. Moreover, 60% of the RO desalted water cost is
149 attributed to energy demand, feed water pre-treatment, membrane cleaning, and eventual
150 membrane exchange [6]. Therefore, recent advances in brackish water RO desalination have
151 mainly centred on membrane materials and process optimisation for reduced water cost and
152 increased water quality.

153 ### 3.1.1. Recent advances in RO membrane materials

154 The semi-permeable membrane is the core of the RO desalination process and directly
155 controls the process production capacity, desalted water quality, energy consumption, and
156 hence the overall efficiency. Thus, attempts to improve the RO process efficiency have centred
157 on enhancing the RO membrane performances such as water permeability, contaminants
158 rejection, and fouling resistance. Commercial RO membranes are categorised into two groups:
159 cellulose acetate (CA) and polyamide thin-film composite (TFC) membranes.

160 CA membranes are produced via phase inversion in which cellulose acetate is precipitated
161 from a polymer solution to form the membrane. Thus, recent advances in the fabrication of CA
162 membranes focus on tailoring the phase inversion process or modifying the membrane surface
163 [25-32]. For examples, Choi et al. [26] optimised the synthesis conditions (e.g. polymer
164 concentration, solvent ratio, and evaporation time) and added multi-walled carbon nanotubes
165 into the phase inversion process to tailor the CA membrane selectivity and permeability.
166 Waheed et al. [27] blended antibacterial chitosan into the dope solution prior to casting the CA
167 membrane. The resultant chitosan-blended CA membrane demonstrated noticeable
168 improvement in antibacterial properties and salt rejection compared to the bare CA membrane
169 [27]. Abedini et al. [32] incorporated TiO_2 nanoparticles into a CA membrane and investigated
170 the impacts of nanoparticles addition on the membrane morphology and thermal stability. The
171 experimental analyses proved that TiO_2 nanoparticles were uniformly dispersed into the
172 membrane structure and increased the membrane porosity, thus improving the thermal stability
173 and water permeability of the CA/TiO_2 membrane [32]. In another study, Yu et al. [29] modified
174 the structure and surface of an original CA membrane via hydrolysis and carboxymethylation.
175 The modification increased the membrane pore size and surface hydrophilicity, hence

enhancing the membrane water permeability. It also rendered the membrane surface more negatively charged, thus improving the membrane salt rejection due to the enhanced Donnan effect as a result of increased membrane surface negative charge [29].

Despite the great efforts to improve their properties, CA membranes have been progressively replaced by the polyamide TFC membranes for brackish water and seawater desalination. Intrinsic drawbacks of CA membranes, including narrow operating pH and vulnerability to microbial attack, restrict the application of CA membranes to desalination of light-load saline water feeds. The polyamide TFC membranes have been dominating the brackish water and seawater desalination markets, and this trend is hardly to change in the foreseeable future [25].

The polyamide TFC membranes are composed of a polyamide active layer laminated on a polysufone substrate via an interfacial polymerisation (IP) process [24]. The polyamide active layer is responsible for salt-water separation while the support layer offers the mechanical strength to the membrane. Compared to CA membranes, polyamide TFC membranes exhibit much higher water permeability and are more resistant to bacterial degradation and hydrolysis, and hence compatible with wider pH feed waters. The layered construction of the polyamide TFC membrane allows for the separate optimisation of the active and the support layer to tailor the performance and durability of the membrane [24]. However, the polyamide TFC membranes are susceptible to the attack of free chlorine in the feed water and more susceptible to membrane fouling than CA membranes [21, 25].

The most notable advance in TFC membranes is the incorporation of nanoparticles into the IP process to improve their desalination efficiency and fouling resistance. Nanoparticles proposed for improved RO membranes include but are not limited to silica [33-35], zeolite [36-38], bentonite [39], metal-organic frameworks (MOFs) [40-42], carbon nanotubes [43, 44], and carbon quantum dots [45, 46]. Given their hydrophilic nature, the incorporation of these nanoparticles into the RO membrane helps enhance the membrane hydrophilicity and facilitate the water diffusion through the membrane, hence increasing the membrane water permeability. The nanoparticles also render the membrane surface smoother; therefore, they increase the fouling resistance of the membrane. For example, the brackish water desalination RO process using a zeolite nanoparticles/polyamide TFC membrane achieved a two-time increase in water flux and salt rejection of 98.4% [36]. The MOFs/TFC membrane exhibited significantly increased water flux and salt rejection (i.e. 41 $L/m^2 \cdot h$ and 97%, respectively) compared to those of the bare TFC membrane (i.e. 30 $L/m^2 \cdot h$ and 69%) when being tested with a brackish water feed [40]. The carbon quantum dots/TFC hollow fiber membrane increased its water permeability by 47% while remaining its high salt rejection of 98.6% [46]. Thus, nanoparticle-

incorporated TFC membranes are deemed the next generation of high performance RO membranes [36]. However, there exist several challenges to commercial nanoparticles/TFC RO membranes including their scale-up difficulty and the high cost together with health and safety issues associated with the use of nanoparticles [47].

Improving the membrane rejection against boron is essential to brackish water RO desalination. The boron rejection of the TFC membranes can be enhanced by regulating the IP process to optimise the polyamide layer. Hu et al. [48] proposed a novel TFC membrane with significantly increased boron rejection achieved by replacing m-phenylenediamine (MPD) with a new sulfonated monomer during the IP process [48]. The novel membrane had a unique membrane surface structure with charge-aggregate induced cavities and alternating hydrophilic-hydrophobic-hydrophilic monomeric structure; therefore, it displayed excellent boron rejection while maintaining an acceptable water flux [48]. Alternatively, La et al. [49] added an aromatic polyamide layer onto a conventional polyamide layer to increase the surface hydrophobicity. The modified TFC membrane achieved a higher boron rejection but at the expense of declined water permeability [49].

Great efforts have also been devoted to surface modification of the polyamide layer for enhanced membrane fouling resistance [50-55]. Most recently, Zhang et al. [50] immobilised positively charged quaternary ammonium groups from 2,3-epoxypropyl ammonium chloride on the polyamide membrane surface to improve membrane fouling resistance and salt rejection. Zhang et al. [55] coated sulfonate polyvinyl alcohol (SPVA) to increase cross-links in the polyamide layer. The experimental investigations demonstrated that fouling resistance together with salt rejection of the polyamide membrane was considerably improved. In a membrane fouling test with a feed water containing 2,000 ppm bovine serum albumin or dodecyl trimethyl ammonium bromide, the SPVA-modified membrane lost only 8% of its initial water flux after 12-hour filtration compared to 28% water flux loss of the virgin membrane [55]. Nevertheless, coating SPVA on the polyamide membrane surface also led to increase in membrane thickness, hence reducing the membrane water permeability. Therefore, the SPVA-surface coated polyamide membranes might be ideal for RO desalination of brackish waters with high fouling propensity whereby a low water flux is reasonably acceptable.

3.1.2. Recent advances in the RO process

Together with the achievements in membrane materials, advances in the RO process have underpinned the growth of brackish water and seawater desalination industries. These technological advances have resulted in marked increase in energy efficiency, water flux, salt rejection, and membrane fouling resistance, thus reducing the cost of RO desalted water. Indeed,

the cost of RO desalted water has been reduced to as low as 0.26 US$/m^3 for brackish water and 0.45 US$/m^3 for seawater desalination [23]. As a result, RO has become the leading process for seawater and brackish desalination applications [4, 23, 56].

Compared to seawater, brackish water largely has lower TDS but higher suspended solids content. Thus, the brackish water RO desalination process is operated at higher water flux and water recovery and under a lower hydrostatic pressure than seawater RO [57-59] (Table 1). However, the brackish water RO desalination process is more prone to membrane fouling than seawater RO. Recent advances in the brackish water RO desalination process have centred on optimising the feed water pre-treatment and process arrangement to mitigate membrane fouling and reduce energy consumption.

Table 1 A comparison between the RO process desalination of seawater and brackish water [22].

Parameters	Seawater RO	Brackish water RO
Water flux (L/m^2·h)	12–17	12–45
Hydrostatic pressure (kPa)	5,500–8,000	600–3,000
Water recovery (%)	35–45	75–90
Salt rejection (%)	99.4–99.7	95–99

Most brackish water RO desalination plants rely on membrane-based pre-treatment to provide quality feed water to the RO membrane modules. Membrane-based pre-treatment combines a pressure-driven membrane filtration process (e.g. MF and UF) with the conventional pre-treatment, adding one barrier against colloids and suspended particles prior to the RO membrane modules. The conventional pre-treatment is only effective to particles larger than 10 μm, whereas the UF process can remove colloids and particles with sizes ≥ 0.1 μm [7]. Therefore, the combined conventional pre-treatment/UF is the most widely used pre-treatment method for brackish water RO desalination plants worldwide [7, 60, 61]. The membrane-based pre-treatment helps improve water flux, increase water recovery, and extend membrane lifetime; however, it also entails increase in capital costs of the brackish water RO desalination process.

Recently, gravity driven membrane (GDM) has been explored as an energy-saving pre-treatment for the RO desalination process [62, 63]. In GDM pre-treatment, the filtrated brackish water after media filtration is dead-end filtered through a UF membrane (Fig. 2). The UF process exploits the gravity to transfer water through the membrane, thus obviating the need for a high-pressure pump as required in a normal UF operation. During the gravity driven UF process, organic compounds (i.e. colloidal particles and particulate organic matter) and the added beneficial eukaryotic organisms in the feed water accumulate and form biofilm on the

membrane surface. The eukaryotic organisms biodegrade organic compounds in the biofilm, rendering the biofilm a porous and heterogeneous structure. As a result, the gravity driven UF process could achieve stable water flux for an extended filtration period of over 100 days without the need for membrane cleaning [62, 63]. Therefore, gravity driven UF pre-treatment not only mitigates membrane fouling propensity but also helps reduce the energy consumption of the brackish water RO desalination process.

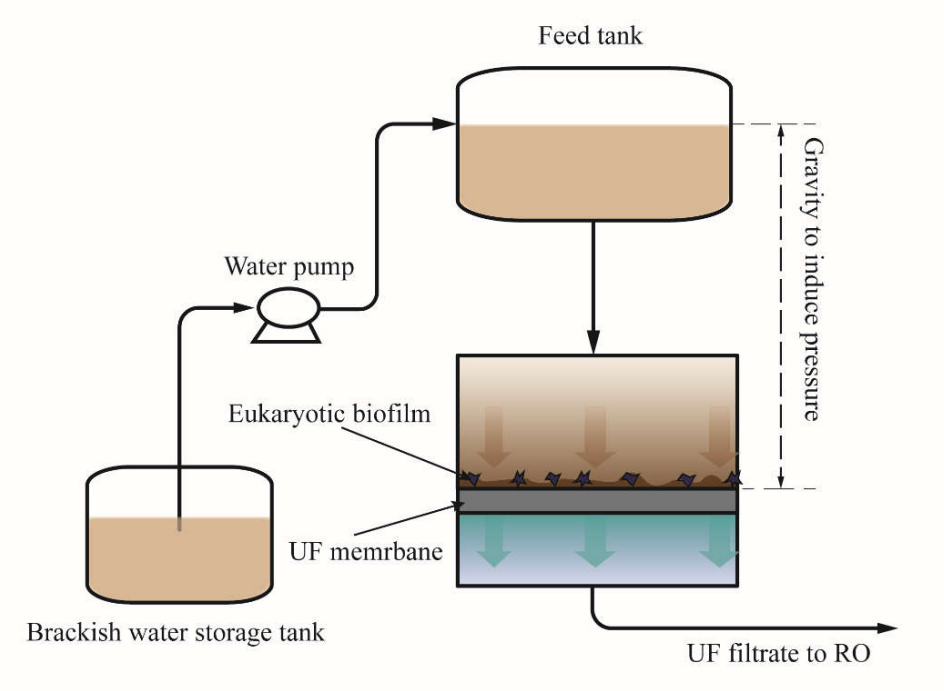

Fig. 2 A schematic describing the gravity driven UF process for brackish water pre-treatment.

Optimising process arrangement plays a vital role in increasing the efficiency of the brackish water RO desalination process. Membranes destined for brackish water RO desalination have a looser polyamide active layer and hence exhibit higher water permeability compared to seawater RO membranes [22]. Moreover, the negative influence of feed water concentration increase on water flux of brackish water RO desalination process is less severe than that observed with seawater RO. As a result, brackish water RO desalination plants are operated at higher water flux and increased water recoveries [12, 22, 64]. Operating the brackish water RO desalination plants at high water recoveries helps utilise the pre-treated water feed more and enhance the energy efficiency of the plants. For example, increasing water recovery from 80% to 93% reduces the brackish water RO desalination energy consumption by 16% [58]. However, brackish water RO desalination plants at increased water recoveries entail higher risks of membrane fouling/scaling and increased process complexity.

A practical approach to increasing water recoveries of brackish water desalination is to combine RO with other desalination processes. For example, RO was coupled with membrane distillation (MD) for increased water recovery treatment of brackish produced water from coal seam gas exploration [65-67]. The brackish produced water was first treated by RO, and the brine following the RO process was fed to the MD process. Given its lower membrane fouling propensity, the MD process reduced the RO brine volume by five folds, resulting in the overall water recovery of 95% for the combined RO/MD process [65-67]. The MD process also concentrated sodium bicarbonate in the RO brine up to its saturation, facilitating its conversion to sodium hydroxide in a subsequent membrane electrolysis process (Fig. 3) [65]. With the availability of solar energy, the combined RO/MD process could offer a technically and economically feasible treatment for brackish produced water from coal seam gas exploration [66].

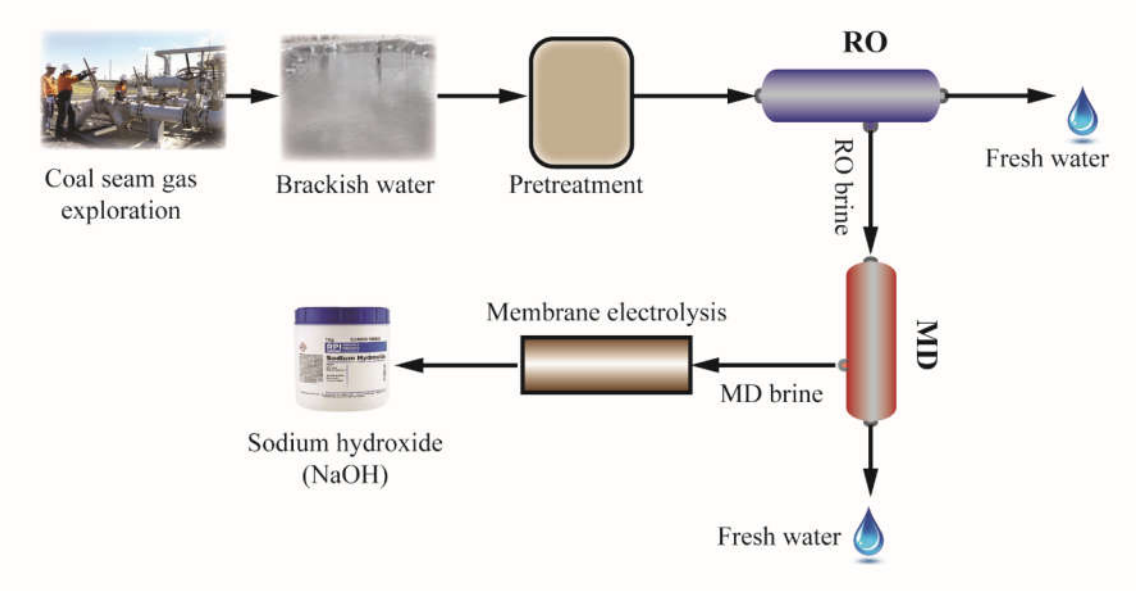

Fig. 3 The combined RO/MD process for zero-liquid discharge treatment of brackish water produced from coal seam gas exploration. The combined RO/MD process brings the sodium bicarbonate concentration in the brackish water up to its saturation limit, thus facilitating its conversion to sodium hydroxide in a membrane electrolysis process.

3.2. Forward osmosis (FO)

Forward osmosis (FO) is an emerging desalination technology whereby an osmotic pressure difference generated by a chemical concentration gradient drives water transport across a semi-permeable membrane. Unlike in RO, there is no external pressure requirement in FO as the transmembrane pressure difference is created by the high osmotic pressure draw solution. For the net flow of water to occur, the osmotic pressure of the draw solution must exceed that of the

feed solution. Therefore, FO membranes allow the selective diffusion of water from a feed solution towards the draw solution, resulting in a concentrated feed stream and a diluted draw solution. In most circumstances, FO desalination involves a two-step process: (1) dilution of the draw solution using FO and (2) fresh water recovery from the diluted draw solution using another desalination process (Fig. 4). Hence, the type of draw solution and the fresh water recovery technique have significant influence on the FO desalination process performance.

For brackish water desalination applications, previous studies have demonstrated that FO provides less severe fouling [60, 68-70] and enhanced water recovery [72, 73] compared with conventional RO technology. However, challenges related to the development of suitable membranes, draw solutions, and the integration of fresh water recovery processes have hindered the uptake of FO processes in desalination markets.

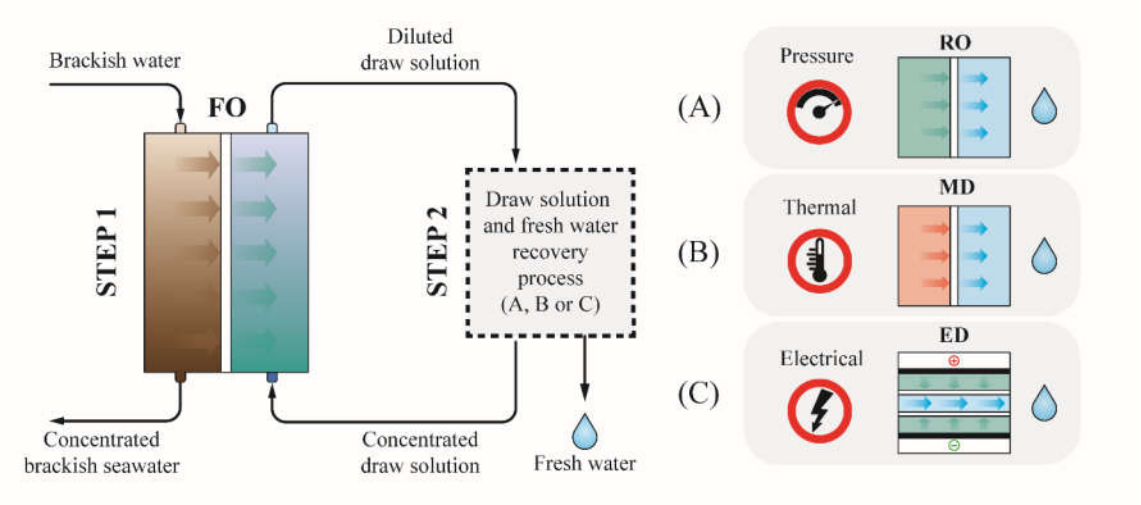

Fig. 4 The two-step FO process for brackish water desalination with (1) osmotic dilution of the draw solution and (2) fresh water recovery from the diluted draw solution using another desalination process.

3.2.1. Recent advances in FO membrane materials

FO membranes are similar to RO membranes as they generally have an asymmetric structure and are composed of an active and support layer. The active layer has a dense selective structure and the support layer is porous to provide mechanical support. The first commercially available FO membrane from Hydration Technologies Innovation (HTI) was a cellulose triacetate (CTA) membrane with an embedded polyester screen or non-woven support. The membrane exhibited reasonable resistance to thermal, chemical, and biological degradation, as well as high mechanical strength [74]. The biggest disadvantage of CTA FO membranes is the thickness of the support layer which limits the membrane flux performance. The development of thin film composite (TFC) FO membranes consisting of a polyamide active layer and a polysufone

support layer has drastically improved the attainable water flux, salt rejection, and chemical resistance compared with CTA membranes [75]. The TFC membrane also provides a wider pH tolerance, but its mechanical stability still requires further improvement [74].

The major challenge associated with FO membranes is the occurrence of internal concentration polarisation (ICP) in the support layer, which reduces the effective osmotic driving force and hence water permeation. ICP is influenced by the thickness, porosity and tortuosity of the support layer [76]. Therefore, advances in membrane fabrication aiming to minimize the negative effects of ICP have focussed on altering the support layer structure. The inclusion of hydrophilic inorganic modifiers into the porous substrate has been extensively studied to improve the support layer characteristics. Carbon-based materials [77-79], titanium dioxide (TiO_2) [80, 81], and zeolite nanoparticle [82] modifiers have resulted in improved water flux and less ICP, attributed to the higher hydrophilicity and porosity of the support layer. For example, a nanofibrous composite membrane containing a scaffold-like nanofiber support layer was developed for brackish water treatment [83]. The nanofibers created a thin support layer with low tortuosity and high porosity, contributing to a reduction in ICP and attaining a higher water flux compared with CTA and traditional TFC membranes. However, the improved water flux was coupled with high reverse salt flux, which is an inherent trade-off for high permeability FO membranes [84]. Alternatively, biomimetic aquaporin-based FO membranes have emerged as a possible game-changer in membrane development due to their exceptional process performance (Fig. 5). Aquaporin membranes are fabricated using aquaporin proteins that provide highly selective water channels [85-87]. Unlike traditional dense polymeric membranes, aquaporin membranes are capable of improving both water permeability and selectivity, without impacting on mechanical strength [85, 86]. Demonstrations of aquaporin and other modified FO membranes to brackish water applications are limited but will increase as improvements are made to fabrication methods and commercial availability.

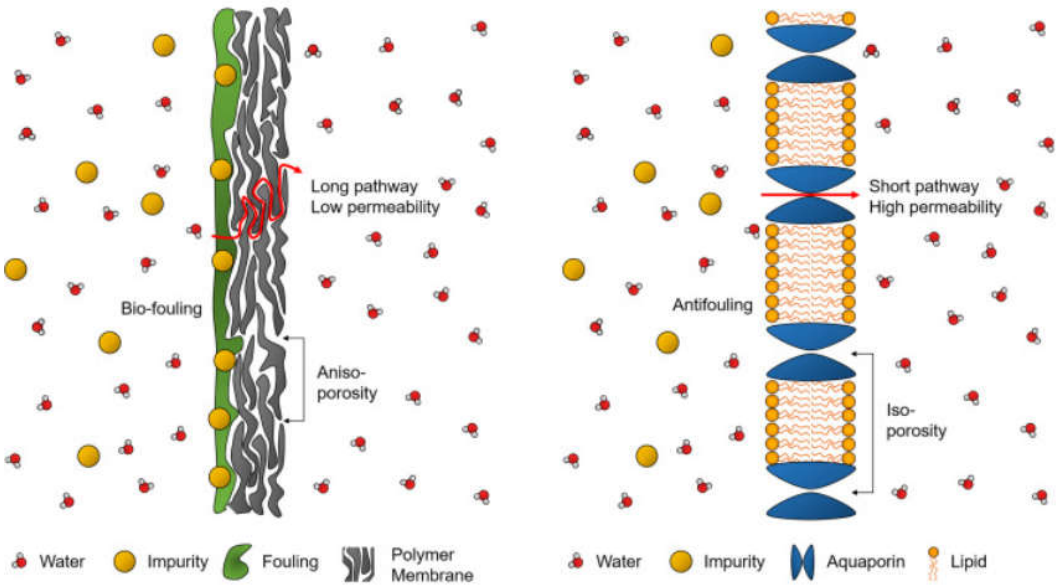

Fig. 5 Illustration for the advantages of aquaporin FO membrane over the conventional TFC FO membrane (with the courtesy from [86]).

Improving the fouling resistance of FO membranes is another integral step for the commercial realisation of FO technology. Although FO is considered to have a low fouling propensity, fouling remains an issue for high fouling potential feed waters such as brackish waters containing high levels of colloids, micro-organisms, organic matter, and minerals [88]. Fouling during FO can lead to deterioration of the membrane and therefore diminish water permeation and separation performance. A number of anti-fouling membranes have been developed and involved chemically modifying the polyamide active layer to increase the membrane hydrophilicity, hence fouling resistance [89]. Modification can be achieved via coating, grafting, or chemical incorporation of hydrophilic materials on or within the membrane surface [90-92]. For example, layered double hydroxides have been incorporated into a polyamide TFC membrane and resulted in a significant resistance to both fouling and chlorine degradation [76]. Additionally, a membrane grafted with polyamidoamine dendrimer for improved ammonia rejection, consequently displayed a strong antifouling performance owing to the surface hydrophilicity and modified surface potential [92]. Despite these promising developments, several challenges remain, and further work is needed to improve membrane performance, anti-fouling capacity, and stability, as well as simplify and develop cost effective fabrication methods.

3.2.2. Recent advances in the FO process

Currently, FO is not comparable to RO for desalination of brackish water for fresh water supply with respect to energy consumption. The FO process alone might consume less energy

than RO; however, the two-step FO brackish desalination process is not considered an energy-saving alternative to RO. Indeed, the energy requirement of fresh water recovery from the diluted FO draw solution greatly exceeds that of the brackish water RO desalination process. As a result, most recent advances in brackish water FO desalination are to strategically develop the one-step FO process or hybridise FO with other desalination processes for increased water recovery and fouling mitigation.

The one-step fertiliser drawn FO (FDFO) concept has emerged as a promising option for low energy brackish water treatment intended for irrigation [93]. FDFO utilises a fertiliser draw solution to simultaneously recover fresh water from brackish water and produce a diluted fertiliser for potential use in agriculture. A number of draw solutions have been evaluated such as potassium chloride amongst other pure and blended fertilisers and proved to be suitable for brackish water treatment [94]. FDFO is a stand-alone FO process whereby no fresh water recovery/draw solution regeneration process is generally needed. Therefore, it is considered to be a low-energy process as the driving force for FO water permeation is provided by the natural osmotic pressure gradient between solutions [95]. However, regeneration of the fertiliser draw solution is a useful step to optimise osmotic pressure and to regulate fertiliser concentrations in the product water. NF has been integrated with FDFO for this purpose and has improved the viability of system scale-up via flux enhancement [68, 96]. Furthermore, evaluation of an FDFO-NF system resulted in a reduced energy consumption of 21% compared with a UF-RO system [97]. Nonetheless, improvements in process flux, reverse solute flux, and membrane cost are essential steps to further improve the commercial viability of FDFO.

Other hybrid FO-desalination processes have been developed for improved water recovery and fouling management for brackish water desalination. Most of these applications employ FO as a pre-treatment step for traditional desalination processes, taking advantage of FO low fouling propensity. For example, FO coupled with NF for brackish water desalination showed less flux decline caused by membrane fouling, and higher amounts of reversible FO fouling [68, 70]. Similar findings have been reported for FO-RO systems [60], even when treating brackish waters with high scaling potential [69]. Another advantage related to the reduced fouling tendency of FO is the capability to increase system water recovery, hence reduce brine discharge. An NF-FO-RO hybrid system achieved >90% simulated water recovery for inland brackish water desalination at TDS between 1,000–2,400 mg/L [69]. Furthermore, an FO-MD hybrid process achieved 81% recovery when treating brackish water RO brine (i.e. TDS = 7,500–17,500 mg/L) [73]. Overall, the high energy consumption of the water recovery process in FO hybrid systems dominates the overall operating costs, whilst the large amounts of FO membrane required for adequate water production represent the major capital cost. Therefore,

there are significant opportunities for thermally driven processes that can utilise low-cost waste heat or solar energy for brackish water desalination [71].

3.3. Electrodialysis (ED)

Electrodialysis (ED) is a well-established electrically driven membrane process with various industrial applications including brackish water desalination for potable water production [98, 99]. The ED process uses ion-exchange membranes and an electric field to desalt saline waters. In an ED cell, cation-exchange membranes and anion-exchange membranes are alternatively arranged between an anode and a cathode to form different compartments as demonstrated in Fig. 6. Given their electrically charged functional groups, cation- or anion-exchange membranes only allow for the permeation of cations or anions, respectively. When a voltage is applied between the cathode and anode, cations and anions in the saline water streams selectively migrate through the membranes, leading to a salt concentration drop in the feed streams but increased salt concentration in the brine streams. As a result, fresh water together with concentrated brine is obtained following the ED process (Fig. 6).

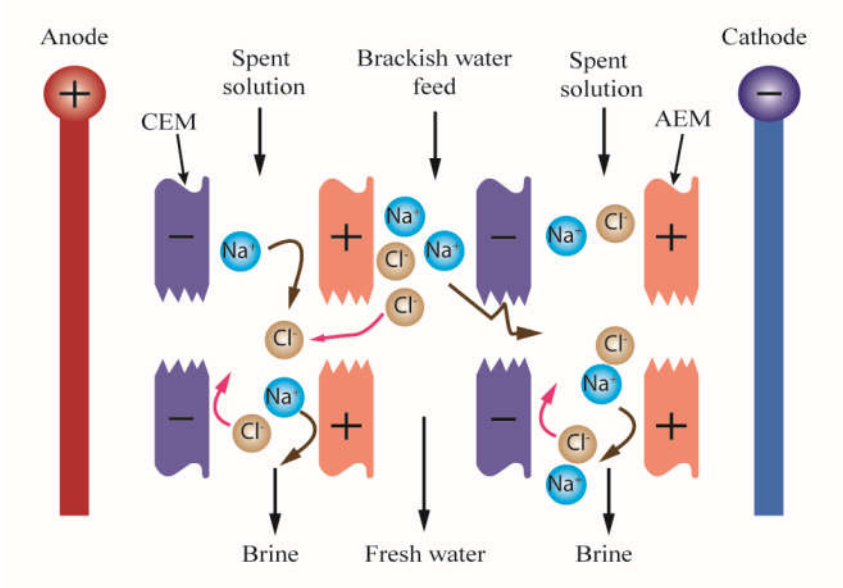

Fig. 6 The working principle of the ED process for desalination of brackish water (adapted from [100]).

Unlike other membrane processes (e.g. RO, FO, and MD), the ED desalination process produces fresh water by removing salts rather than water from saline waters. Because salt concentration in brackish water is negligible compared to water concentration, brackish desalination by removing salts is much more energy efficient compared to removing water. Thus, for brackish water desalination the ED process exhibits an overwhelming advantage over other membrane processes with respects to energy efficiency [21]. Indeed, the brackish water

ED desalination process exhibits a significantly lower specific energy consumption (i.e. 0.7–2.5 kWh/m^3) compared to the RO process [2, 101].

The desalination performance of the ED process is critically dependent on the transport rate of ions through the ion-exchange membranes. Elevated ions transport rate will result in decreased salt concentrations and increased flowrate of the fresh water stream, thus improving the fresh water quality and production rate of the ED process. The ions transport rate through the ED membranes is regulated by ED membrane characteristics and applied current [102].

As an electrically driven desalination process, electricity is the primary energy input of ED. The electricity consumption is mainly for electric field between the cathode and anode, and it increases with the voltage drop over the electrodes. In turn, the voltage drop over the electrodes is attributed to the overall Ohmic resistance and the non-Ohmic voltage drop [102]. These two parameters are dependent on salt concentrations in the brine and fresh water compartments. Increased salt concentration gradient between the brine and fresh water compartments elevates the non-Ohmic voltage drop, whereas the overall Ohmic resistance decreases when the salt concentrations in compartments increase. The overall Ohmic resistance is also greatly affected by the ion exchange membrane properties including the membrane ionic conductivity [102].

Another electrically driven process that has gained increasing attention for desalination of brackish water is capacitive deionisation (CDI) [103-105]. Like ED, the CDI process employs an electrical field to drive the movement of ions towards electrodes to achieve the desalination of brackish water. However, unlike ED, CDI is strictly not a membrane-based desalination process because it does not require a membrane for salt-water separation. Instead, the CDI process employs porous electrodes to adsorb ions from the brackish water feed and hence desalinate it. The porous electrodes are the core of the CDI process as they profoundly affect the process efficiency. A key property of the CDI electrodes is their specific capacitance, which is measured in Faraday per gram (F/g) [100]. Specific capacitance indicates the electrostatic adsorption capacity of electrodes. In other words, electrodes with higher specific capacitance can adsorb more salt ions from the feed water and hence are compatible with brackish water feeds with higher salinity. Most current electrodes used for the CDI process have specific capacitance below 125 F/g [106]. As a result, the application of the CDI process has been limited to the desalination of brackish water with salinity less than 2,000 mg/L [107, 108], which is much lower than the feed water salinity allowed for the ED desalination process.

3.3.1. Recent advances in ED membrane materials

In ion-exchange membranes, the charged functional groups attached to a polymer matrix are responsible for the selective permeation of ions through the membranes. The negatively

charged groups on the cation-exchange membranes exclude anions, thus rendering the membranes preferentially permeable to cations. Likewise, the anion-exchange membranes are selectively permeable to anions due to their positively charged groups. Key properties of ion-exchange membranes that control the desalination efficiency of the ED process are ionic conductivity, perm-selectivity, and chemical, thermal, and mechanical stability.

Given the maturity of the ED process, there have been relatively limited recent advances in ion-exchange membrane materials. Only few studies on ion-exchange membranes have recently been reported [109-112]. Most notably, Shukla and Shahi [109] fabricated a novel composite cation-exchange membrane whereby imidized graphene oxide with multi-functionalized groups was incorporated into the sulfonated polymer matrix to increase the ionic conductivity, per-selectivity, ion-exchange capacity, and stability of the membrane. Afsar et al. [111] fabricated a cation-exchange membrane with integrated cationic and anionic layers for increased membrane perm-selectivity. A carboxyl membrane base was prepared from polyvinyl alcohol (PVA) and subsequently coated with the cationic and anionic layer of quaternized poly phenylene oxide and sulfone poly phenylene oxide, respectively. The double-layer structure increased the per-selective behaviour of the fabricated cation-exchange membrane, rendering it highly promising for desalination of brackish water containing monovalent and divalent cations [111].

Like other membrane processes, membrane fouling is a challenge to the application of ED for brackish water desalination. There is a consensus that brackish water contains high contents of negatively charged organic compounds. In the ED process, when the electric field is applied, these organic compounds (i.e. in the forms of colloidal particles and particulate organic matter) move toward the anode but are retained and subsequently deposited on or within the anion-exchange membrane surface. The deposited colloidal layers reduce the ionic conductivity and negatively alter ion selectivity of membrane, hence deteriorating the desalination efficiency of the ED process. To increase the fouling resistance of anion-exchange membranes against negatively charged colloidal particles, Mulyati et al. [113] and Vaselbehagh et al. [114] incorporated high molecular mass surfactants into the anion-exchange membranes to promote their negative surface charge density, hydrophilicity, and roughness. The experimental results confirmed that the surface-modified anion-exchange membranes were not only more resistance to fouling but also more chemically and mechanically stable than the original membranes. Pre-treatment of the brackish water feed using membrane filtration processes (e.g. MF and UF) is also applied to mitigate membrane fouling and simultaneously improve organic compounds removal of the brackish water ED desalination process [102].

3.3.2. Recent advances in the ED process

Pre-treatment of brackish water has been also practised to prevent membrane scaling caused by sparingly soluble inorganic salts (i.e. $CaCO_3$ and $CaSO_4$) during the brackish water ED desalination process at high water recovery [115, 116]. The addition of anti-scalants to the brackish water feed can effectively prevent the deposition of $CaSO_4$ on the membrane surface; however, it is unworkable for $CaCO_3$. Recently, Sayadi et al. [115] experimentally assessed the efficiency of three physical membrane scaling prevention methods with $CaCO_3$ using magnetic, ultrasonic, and pulsed electric field during the ED desalination process. These physical anti-scale treatments facilitated the homogeneous formation of $CaCO_3$ in the solution but no on the membrane surface, thus effectively preventing membrane scaling [115]. The results obtained from this lab-scale testing are promising; nevertheless, many further works are required before these anti-scale methods can be practically applied for brackish water ED desalination.

A breakthrough in membrane scaling prevention in ED desalination is the introduction of the electrodialysis reversal (EDR) concept [102, 117, 118]. Indeed, EDR can be considered an enhanced anti-fouling ED process. During the EDR operation, the migration of ions and organic matter across the ED cells is regularly reversed by switching the electrode polarities and the diluate and brine channels. The foulants deposited on the membrane surfaces in the previous ED cycle are detached and released into the brine streams before being rinsed out of the ED cells. The fouled membranes are effectively self-cleaned during the EDR operation. Therefore, the EDR process is remarkedly more resistant to membrane fouling and hence offers a more cost-effective desalination means for brackish water than the ED process. Due to its reduced membrane fouling/scaling tendency, the EDR desalination process can be operated at higher water recovery than the ED process [117]. Given these considerable advantages, the EDR process is rapidly gaining its popularity. Nevertheless, as concluded in [118], there is still a gap between lab-scale testing and full-scale industrial applications, and hence more pilot-scale studies are required to scale up the EDR process.

3.4. Membrane distillation (MD)

The thermally driven membrane distillation (MD) process has considerable potentials for desalination of brackish water. The MD process uses a hydrophobic, microporous membrane to separate a brackish water feed and a fresh distillate stream. Given its hydrophobic nature, the MD membrane allows only the permeation of water vapor but not liquid water. Therefore, in theory the brackish MD desalination process can achieve a 100% salt rejection to produce pure distillate [119, 120]. Moreover, the MD process is significantly less susceptible to fouling than other membrane processes (e.g. RO, FO, and ED) due to the discontinuity of liquid water across

551 the MD membrane [119, 120]. The low membrane fouling tendency might reduce feed water
552 pre-treatment before the brackish water MD desalination process. More importantly, the MD
553 process is driven by the temperature difference across the membrane, and thermal energy is the
554 primary process energy input. Thus, low-grade heat sources such as waste heat or solar thermal
555 energy can be explored to meet the energy demand and hence to reduce the energy costs of the
556 brackish water MD desalination process.

557 Non-wetting of the membrane pores is a critical condition for the MD process. The
558 membrane pores remain dry when the hydrostatic pressure of the process streams is limited
559 below the liquid entry pressure (LEP) [120-125]. When the LEP is exceeded, liquid water can
560 penetrate and render the membrane pores wetted. Consequently, liquid water (i.e. hence
561 dissolved salts and contaminants) rather than water vapor transfers through the membrane,
562 hence leading to deterioration in the MD process salt-rejection [121-124]. Membrane pore
563 wetting also leads to decline in the MD water flux because of the decreased active membrane
564 surface area for water evaporation. LEP is dependent on the membrane properties and solution
565 characteristics as expressed below [120]:

566 $$LEP = \frac{-2B\lambda_L \cos\theta}{r_{max}} \quad (2)$$

567 where B is the geometric factor representing pore structure, γ_L is the liquid surface tension, θ
568 is the liquid-membrane contact angle representing the membrane hydrophobicity, and r_{max} is
569 the maximum membrane pore radius.

570 The most notable factor that might induce membrane wetting in MD desalination of brackish
571 water is the varied particulate matter content. Organic compounds and surfactants in brackish
572 water reduce the solution surface tension and might attach to the membrane and subsequently
573 alter membrane surface hydrophobicity. Thus, they reduce the LEP value and increase the risk
574 of membrane pore wetting. Indeed, LEP linearly decreases with the increased organic solutes
575 concentration in the solution [120]. As a result, it is critical to remove organic compounds and
576 surfactants from the brackish water prior to the MD process to prevent membrane pore wetting.

577 Thermal energy efficiency is key aspect of the brackish water MD desalination process. The
578 MD process thermal energy efficiency is evaluated using two parameters: specific thermal
579 energy consumption (STEC) and gained output ratio (GOR). While STEC directly shows the
580 amount of thermal energy demand (i.e. in kWh) to produce one volumetric unit (i.e. in m^3) of
581 distillate, GOR demonstrates the heat recovery efficiency of the MD process [120].

3.4.1. Recent advances in MD membrane materials

Like in other membrane processes, in MD the membrane exerts strong effects on the process performance. The membrane properties regulate the salt removal as well as the heat and water flux through the membrane. Therefore, the MD membrane decisively influences the desalination efficiency (i.e. distillate quality and production) and the energy consumption of the MD process.

Most recent advances in MD membranes are orientated toward improving their water flux and wetting resistance. For example, a large number of attempts have tried to fabricate super hydrophobic membranes to enhance the membrane wetting resistance and water flux of the MD process [126-133]. Super hydrophobic MD membranes are achieved by coating nanoparticles [127, 130, 134, 135], highly hydrophobic perfluorinated copolymers [133, 136], or microsphere [128] on the surface of the hydrophobic polymer membrane probably followed by surface modification (i.e. fluorinated modification). The experimental results demonstrate that the MD process using super hydrophobic membrane exhibited significant improvement in both water flux and membrane fouling/wetting resistance [127-130, 133, 134, 136]. For example, the MD process with the anti-wetting super hydrophobic membrane under wetting-intense conditions could noticeably increase water flux from 26.0 to 29.9 $L/m^2 \cdot h$ and delay the membrane wetting occurrence from 40 to 180 minutes [129].

Another approach to improving water flux is to explore multi-layer MD membranes. Most MD systems use commercial MF membranes consisting of a hydrophobic active layer laminated on a hydrophobic support layer with relatively high thickness. Recently, the novel electrospinning method has been deployed to prepare dual hydrophobic-hydrophilic layer or three layer membranes specifically for MD [135, 137-142]. The electrospinning method allows for effective control of membrane layer thickness and pore sizes and the addition of nanoparticles to the membrane layers. The hydrophilic support layer after being wetted by the distillate helps reduce the pathway of water vapor inside the membrane pores (Fig. 7). Therefore, the electrospun multi-layer MD membranes presented noticeably enhanced water permeability and wetting resistance compared to commercially available MD membranes [137-139, 141, 143].

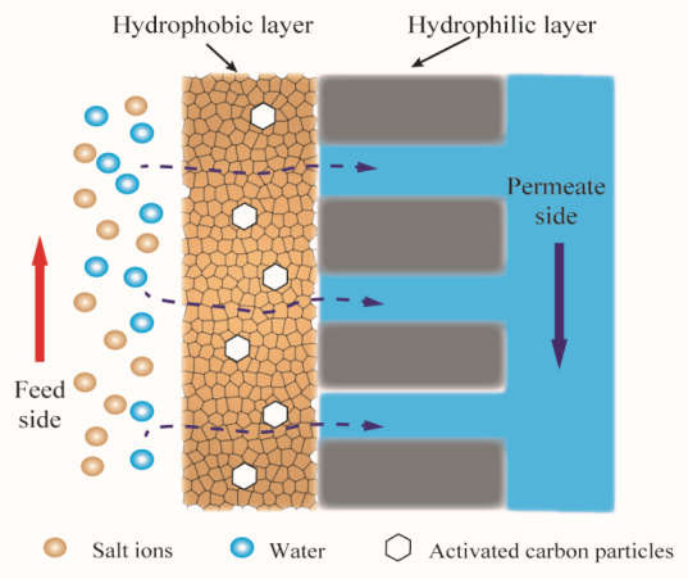

Fig. 7 A schematic of a cross section of a dual hydrophobic-hydrophilic MD membrane prepared using the electrospinning method (adapted from [143]).

Water flux of the MD membrane can also be enhanced by enlarging the membrane pore sizes and increasing membrane porosity. In general, the MD membrane with larger pore sizes and higher porosity exhibits increased water flux and less conductive heat loss but at the expense of reduced mechanical strength and LEP [120, 133]. Given the versatile electrospinning method, MD membranes with large pore sizes and porosity and sufficient mechanical strength and wetting resistance have been obtained by reinforcing the substrate layer [144-146]. These reinforced MD membranes demonstrate great potentials for desalination applications whereby high water flux and membrane wetting resistance are required [144-146].

3.4.2. Recent advances in the MD process

There has been a consensus that MD is an emerging desalination process and it is currently not comparable to RO, FO, and ED for brackish water desalination applications. As a thermally driven process, MD requires huge amount of heating and cooling to achieve fresh water from saline waters. Thus, MD is rarely considered an ideal process for brackish water desalination. However, unlike other membrane desalination processes, MD can be coupled with low-grade heat sources such as waste heat from other industrial processes or solar thermal energy. With the availability of these low-grade heat sources, the energy cost of brackish water MD desalination can be reduced. Alternatively, MD can be combined with other membrane processes for improving the water recovery and energy efficiency of brackish water desalination [65-67].

In practice, most recent pilot MD demonstrations for brackish water or seawater desalination are on solar-powered or waste heat-driven processes [147-155]. This might be attributed to the high process energy consumption of MD, which is currently considered one of the key hurdles for its commercialisation. Coupling MD with solar thermal energy or waste heat helps alleviate its high energy consumption and renders it more competitive for brackish water or seawater desalination. For example, Chaffed et al. [154] developed and experimentally investigated the performance of an integrated solar-driven pilot MD system for potable water production from brackish water. The investigation results confirmed the viability of the solar-driven MD for potable water production from brackish water [154]. Dow et al. [155] assessed membrane fouling propensity of a pilot MD process driven by waste heat from a gas fired power station during a three-month operation [154]. Due to the limited waste heat temperature (i.e. <40 °C), the MD process exhibited a low water flux of 3 $L/m^2 \cdot h$. However, despite testing with the real power station effluent, membrane fouling was only evident at the very end of the operation, and the MD process achieved 99.9% salt rejection throughout the operation [154].

Recently, several novel approaches to enhancing the solar radiation absorption efficiency have been implemented to facilitate the solar-driven MD desalination process [147, 156]. The most notable example is the addition of nanofluids to the MD feed water stream to increase the solar radiation absorption locally at the membrane surface, thus enhancing the utilization of solar radiation and simultaneously obviating the need for the conventional solar thermal collectors [156]. The energy utilization efficiency and water flux of the MD process with added nanofluids were improved both by nearly 60% compared to those of the process without nanofluids [156]. Most importantly, the MD membrane retained 100% of nanofluids, thus producing distillate with excellent quality (i.e. with salinity <10 mg/L) [156]. These novel approaches are promising for the solar-driven MD desalination of brackish water with respects to energy consumption and hence production cost reductions. Nevertheless, further studies are required to elucidate the long-termed effectiveness of these novel heating methods.

Conclusions

Brackish water has become a viable source to augment fresh water supply. Membrane processes including RO, FO, ED, and MD, have been the key technology for brackish water desalination. Widely varied characteristics of brackish water present considerable challenges to brackish water membrane desalination processes. Therefore, recent advances in membrane materials and process designs of these four membrane processes largely focus on improving fouling resistance, boron rejection, water flux, and energy efficiency. New membrane materials including nanoparticle-incorporated thin-film composite polyamide, aquaporin, and super-

hydrophilic or super-hydrophobic polymers demonstrate great potentials with respects to enhanced water permeability, fouling resistance, and rejection against small-molecule contaminants. Recent innovations in process designs also help facilitate the applications of the membrane processes for brackish water desalination. Pre-treatment using gravity-driven UF effectively prevents membrane fouling and in tandem reduces the energy consumption of brackish water RO desalination. The fertiliser-drawn FO process offers a cost- and energy-effective treatment of brackish water for irrigation. Reversal ED has proved itself an energy-efficient and fouling resistant process for brackish water desalination. Finally, solar-powered or waste heat-driven MD processes achieve quality fresh water from brackish water with markedly reduced energy costs. Recent advances in membrane materials and processes are highly promising and expected to be the game changers for brackish water desalination. However, great efforts are required to boost the scaling up of these novel membrane materials and process designs.

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Conflict of interest statement

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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