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Advances in Forward Osmosis (FO) technology for enhanced efficiency and output: A critical review

D. Dsilva Winfred Rufuss[a,b], V. Kapoor[a], S. Arulvel[a], P.A. Davies[b*]

[a]School of Mechanical Engineering, Vellore Institute of Technology (VIT), Vellore-632014, Tamil Nadu, India

[b]School of Engineering, University of Birmingham, Edgbaston, Birmingham B15 2TT, UK

[*]Corresponding author: P.A. Davies (email: P.A.Davies@bham.ac.uk)

Abstract

Freshwater scarcity is one of the most important issues facing the world today. To address this issue, processes have been developed to purify and desalinate water at an industrial scale, especially based on membrane reverse osmosis RO. However, because of the drawbacks of conventional RO – including the inability to handle high salinity and susceptibility to fouling – forward osmosis (FO) has been introduced as a complementary technology. FO can be coupled with other desalination techniques like membrane distillation and RO to remedy these issues. We aim here to review recent advances in FO and the challenges facing this technology. Important parameters in FO operation include transmembrane water flux and output, energy consumption, fouling, draw solution type and regeneration, and membrane type. Several methods to increase the water flux are discussed, including changes in system temperature, development and alterations in draw solution (DS) properties, modelling and development of new membranes, and techniques to reduce concentration polarization. These developments help to increase water flux and water recovery and to mitigate membrane fouling and concentration polarization. We also discuss the various applications of these novel techniques in different areas, and how they can improve the efficiencies of hybrid systems. Finally, we

make recommendations for future developments, to allow the use of FO at a large scale in water purification systems.

Keywords: Forward osmosis; draw solution; concentration polarization; water flux; water recovery

Nomenclature

AL-DS	Active layer facing draw solution
AL-FS	Active layer facing feed solution
CNT	Carbon Nanotube
CTA	Cellulose Triacetate
DS	Draw Solution
ECP	External Concentration Polarization
FO	Forward Osmosis
FS	Feed Solute/Solution
GO	Graphene Oxide
ICP	Internal Concentration Polarization
LMH	Liter/square meter/hour
M	molarity
MED	Multi Effect Desalination
MSF	Multi Stage Flash
PRO	Pressure Retarded Osmosis
RO	Reverse Osmosis
TFC	Thin-Film Composite
UN	United Nations

1. Water scarcity

Freshwater scarcity is one of the most prevalent global problems, affecting more than two-thirds of the world population (Abedin et al., 2020). Available freshwater represents less than 0.015% of water on Earth (Marvuglia et al., 2020). Freshwater resources are constantly being

depleted and degraded by pollution, global warming, population growth, and increasing rates of consumption accompanying economic development (Omar et al., 2020). Per capita domestic consumption is about 10 times higher in developed countries than in developing countries (World Health Organisation, 2021). If the current trends continue, it is predicted that by 2030, water demand is expected to outstrip supply by 40% (Omar et al., 2020).

About 81% of global wastewater is discharged back into the natural environment without any further treatment or reuse (He et al., 2020). It is often cheaper and easier to source freshwater than it is to treat and recycle wastewater (World Vision, 2018). Around 845 million people lack access to basic drinking water today (World Health Organisation, 2019). Rising freshwater demand has spurred on research and development in the area of desalination in recent years (Sakthivadivel et al., 2021). Desalination refers to the process in which impure saline water is treated by removing the mineral salt content, thus purifying it to within safe drinkable limits (Lu et al., 2020). According to a report by the United Nations (UN), almost 1% of the world relies on desalination technologies to meet their daily needs (Nassrullah et al., 2020). Among the various desalination techniques, membrane osmotic separation processes are being widely adopted to extract impure elements or ions from saline water and produce pure drinking water (Das and Warsinger, 2021).

2. Membrane osmotic separation processes

The two main osmotic separation processes are reverse osmosis (RO) and forward osmosis (FO). RO refers to a water purification technique in which pressure is applied to overcome the osmotic pressure caused by concentration difference of solute across a semi-permeable membrane (Park et al., 2020). This method is used at large scale in water purifiers to remove both dissolved and suspended impurities from water (Touati and Rahaman, 2020). RO has become highly efficient in treating relatively dilute feeds such as brackish water and seawater, but it cannot be used to treat solutions having high concentrations of impurities (X. Li et al.,

2019). For example, hypersaline water has very high osmotic pressure, requiring operating pressures in excess of those RO membranes can withstand. Another limitation of RO is its susceptibility to membrane fouling and scaling (Saleem and Zaidi, 2020).

Over the past decade, there has been much research into the techniques of forward osmosis (FO) as a complementary technique to RO (Chiao et al., 2021). Like RO, FO is an osmotic process, which uses a semi-permeable membrane separating the draw solute/solution (DS, of high concentration) and the feed solute/solution (FS, of lower concentration) (Suwaileh et al., 2020). FO can treat highly concentrated brine with salinity >70000 ppm. Brine treatment is important, not only for harvesting of freshwater, but also for recovery of minerals and for management of brine effluents (Zhu et al., 2020). Unlike RO, no hydraulic pressure is applied in FO; instead, the driving force (i.e. osmotic pressure gradient) is provided by the concentration differences across the membrane (Ibrahim et al., 2018).

Even though FO can treat highly concentrated brine, there are still problems to address before FO can realize its full potential in industrial applications (Mahto et al., 2021). These problems include membrane fouling, concentration polarization, declining output with continuous usage, and difficulties in scaling up (Akhtar et al., 2021). Despite the rapidly growing number of research papers in this field, there is only a limited number of review articles related to the topics of draw solutes (Cai and Hu, 2016), graphene oxide-based membranes (Wu et al., 2020), carbon-based nanomaterials (Yadav et al., 2020), draw solutes regeneration (Luo et al., 2014), fouling mitigation (W. J. Lee et al., 2020) and process modeling (Sekino, 2021). Key topics including efficiency, novel membranes (e.g. silica materials), and the influence of parameters such as temperature and concentration polarization, have not been adequately discussed (Blandin et al., 2020). Moreover, a critical review giving insights into membrane materials, membrane advancement in the active and support layer of FO is timely and undoubtedly necessary to support future membrane development.

In this new review, we discuss the techniques and principles of FO, and we survey recent approaches to enhance water flux and mitigate deficiencies of the system. The review covers optimization of operating temperature, selection of appropriate DS to generate high osmotic pressure, selection of appropriate membranes to reduce membrane fouling and scaling, and selection of suitable membrane configuration and flow velocities to reduce the effects of ECP (external concentration polarization) and of ICP (internal concentration polarization). In addition, we explicitly address the following key questions related to membrane advancements: Which is better, virgin cellulose triacetate (CTA) or thin-film composite (TFC) membrane? What modifications are possible in the active layer and support of FO membranes? What is the role of nanoparticles in membrane modification? Addressing the above questions will help researchers and academics to understand membrane development and advance this topic further, thus widening the range of FO applications. Further, novel industrial applications of the FO process will be discussed here.

3. Overview of the FO system

Before presenting the detailed review, we introduce in this section the main components of the FO system; namely, the feed solution (FS), draw solution (DS), FO membrane, and draw solution regeneration system (Fig. 1).

The feed solution is the water to be treated. Types of feed solution include wastewater from various industries and hypersaline water. The draw solution is more concentrated than the feed, creating an osmotic pressure gradient and inducing water flow from feed to draw solution via the FO membrane (Wang and Liu, 2021). The FO membrane permits only water molecules to pass while rejecting other species. FO rejection rate and reverse solute flux are the two major performance characteristics of a FO membrane (Blandin et al., 2020). FO rejection rate refers to how efficiently a FO membrane prevents the other contaminants from permeating (Blandin et al., 2020); whereas reverse solute flux refers to the rate of draw solute diffusion across the

membrane and into the feed solution (Suwaileh et al., 2020). Ideally, the rejection rate should be high and reverse solute flux should be low (Wang and Liu, 2021). During the FO process, the feed solution gets concentrated and the draw solution gets diluted. So, the draw solution should be further treated with a suitable regeneration system to recover the freshwater (Wang and Liu, 2021). Types of regeneration system include nanofiltration, RO, thermo-responsive regeneration and magnetic regeneration (Luo et al., 2014).

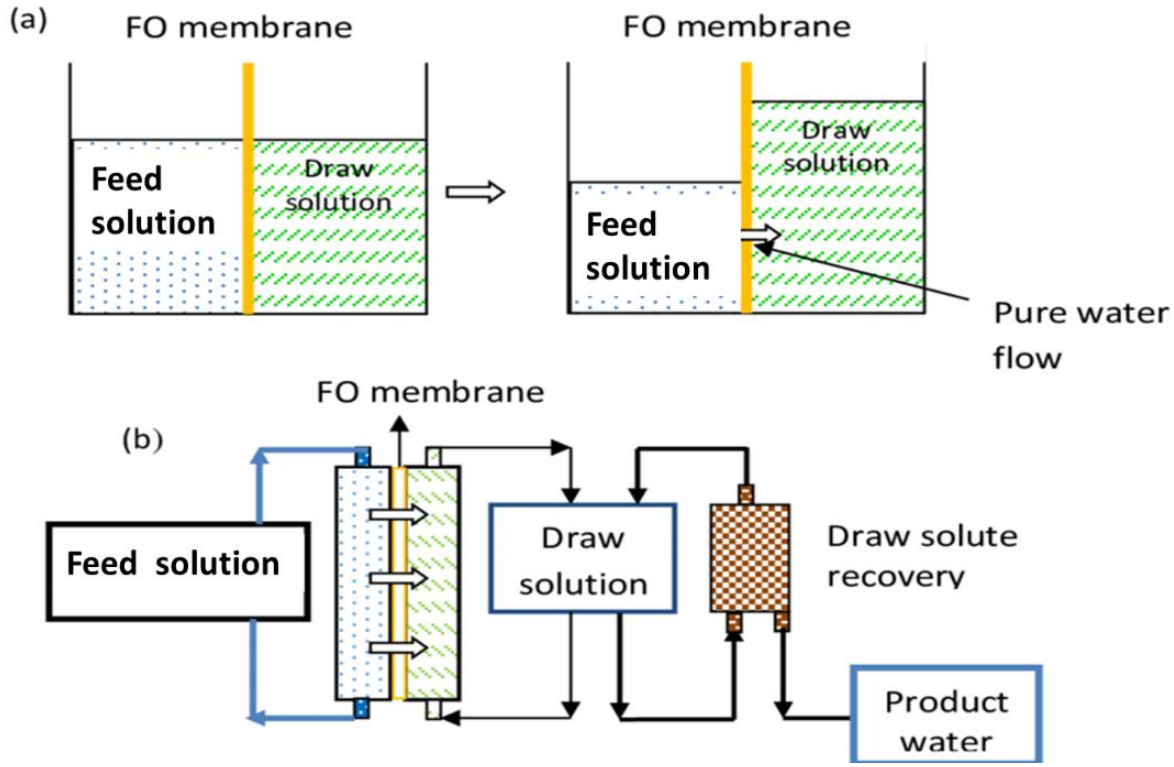

Fig. 1 (a) concept of FO (b) basic components of FO (Hai et al., 2014)

4. FO Membranes

4.1.Cellulose triacetate (CTA) vs thin-film composite (TFC) membranes

FO uses a semipermeable membrane that is made of either cellulose triacetate (CTA) material or thin-film composite (TFC) material (Akther et al., 2015). CTA membranes were the first commercially available membrane. They have advantages of good mechanical properties, hydrophilicity and low fouling (Akther et al., 2015). However, low pH, rejection

rate and permeability were found to be drawbacks of the CTA membranes. To overcome these drawbacks, TFC membranes were developed which can have high pH and high rejection rate (Wang et al., 2018). The rejection of CTA membranes is 85-90%, compared to 90-95% for TFC membranes. This is because of the lower solute permeability and higher water permeability of TFC membranes (Low et al., 2015). For example, CTA membrane has shown solute permeability coefficient of 0.40 LMH (Goh and Ismail, 2018) compared to 0.26 LMH for TFC membrane (Kim et al., 2017); and water permeability of 0.14 L/m^2.h.bar compared to 0.74 L/m^2.h.bar for TFC membrane (Seah et al., 2020).

CTA membranes are mostly used in domestic desalination applications and less concentrated wastewater industrial applications due to their good mechanical strength and lower fouling propensity (Shakeri et al., 2019b). Around 95-99% of trace antibiotics have been removed from the wastewater in seawater desalination industries using these CTA membranes (Liu et al., 2015). In addition, CTA membranes are often used in coal processing industries to dewater waste coal slurry because of good fouling resistance (Lutchmiah et al., 2014). In one trial, CTA membranes were able to endure thirty continuous tests with coal slurry, confirming their mechanical strength (Shakeri et al., 2019b). In summary, CTA membranes can be used to treat wastewater at low concentrations in large volumes.

TFC membranes can treat highly contaminated and concentrated wastewater. They have a wider range of applications, extending to wastewater from beverage industries (Blandin et al., 2020), pharmaceutical industries (Zhou and Lee, 2016), textile and manufacturing industries (Korenak et al., 2019). For example, TFC membranes have been used to remove ammonia and cyanides from industrial waste coke water, respectively, in beverage and pharmaceutical industries with 98% removal efficiency (Blandin et al., 2020). Similarly, they can remove mercury from industrial wastewater in manufacturing and textile industries with 96% removal efficiency (Nguyen et al., 2015).

4.2. Fabrication methods

In general, the fabrication of a FO membrane refers to the process of building the membrane with the polymer and mesh structure needed to provide the desired stability, surface properties and fouling resistance. There are various methods to fabricate the membrane, depending on the type of membrane and polymer being used. These include interfacial polymerisation, layer-by-layer fabrication, phase inversion through immersion precipitation, and electrospinning. Interfacial polymerisation is the most common method. Here the support layer is soaked in an aqueous solution of selected polymer and then immersed in di-isocyanate solution, thus improving the structural morphology of the membrane (Purkait et al., 2018).

In the layer-by-layer technique, positively and negatively charged materials are deposited on top of each other in an alternating fashion to form a sheet (Salehi et al., 2017). Various polymers and nanoparticles, such as graphene oxide and polyethylene glycol, can be coated on this membrane to decrease its structural parameter (Xu et al., 2015). In phase inversion through precipitation, selected thermoplastic polymers including polyacrylonitrile (Tiron et al., 2017), polysulfone (Suwaileh et al., 2018) and polyether sulfone are suspended in a coagulation bath by mixing it with the appropriate solvent to improve the hydrophilicity, mechanical, fouling resistance and thermal stability of the membrane (Xinfeng Zhang et al., 2018).

In electrospinning, an electric force is used to deposit the charged threads of polymer on the surface of the membrane. It is the most commercial method to coat nanofibers or nanoparticles on the surface of a membrane. Electrospun FO membrane has good mechanical stability and is flexible during operation (Nune et al., 2017). In general, phase inversion and electrospinning methods are preferred to fabricate flat-sheet and HF membranes, whereas interfacial polymerisation and layer by layer methods are preferred to deposit polyamide and nano-range particles, respectively, on the support layer of the FO membrane.

4.3. Modifications to CTA membranes

Despite the wide range of applications of CTA membranes (see Section 7), this membrane has limitations of low permeability, susceptibility to different types of fouling, and low salt rejection, making it unsuitable to treat some effluents (Lutchmiah et al., 2014). To overcome these limitations, researchers modified have the active layer and support layer of the FO membrane to enhance the surface morphology, structural stability and membrane characteristics (Suwaileh et al., 2018).

For example, the active layer of the CTA membrane was modified with polydopamine-coated polyethylene glycol to reduce the biofouling (Xu et al., 2019). Similarly, the modification of the support layer using components like polyester, polyethylene terephthalate mesh, polyvinylidene and polyether-sulfone has improved the structural parameter, reduced fouling, and enhanced the hydrophilicity of the virgin CTA membrane (Zhou et al., 2020). For example, polyethylene terephthalate mesh was used in the CTA support layer to enhance the water flux as it reduces the membrane fouling and improves the hydrophilicity of the membrane (Xu et al., 2019).

4.4. Addition of nanoparticles to CTA membrane

To further enhance properties of hydrophilicity, porosity and permeability of CTA membranes, nanoparticles such as functionalized carbon nanotubes (CNT), carboxylic and amine nanofibers, have been used in both active and support layers (Shakeri et al., 2019a). For example, functionalized carbon nanotubes on the membrane selective layer improved the fouling resistance of the membrane and increased the water flux by 50% (Chiao et al., 2019). This is due to the improved hydrophilicity of the membrane modified with CNT. In another instance, silver polydopamine nanoparticles were coated on the membrane active layer reducing the biofouling in the membrane and enhancing the water flux. This was due to the

198 improved anti-biofouling and anti-microbial properties of the modified membrane (Chiao et
199 al., 2019).

200 Increasing the surface area of the support layer can enhance the water flux, permeability
201 and mechanical strength of the virgin CTA membrane. Thus, silica nanoparticles have been
202 used to increase the surface area of the membrane by 15%, increasing permeability seven-fold
203 (Shakeri et al., 2019b). Another example is the use of graphene oxide sheets and zeolites in
204 the support layer to improve the surface area of the FO membrane. These nanoparticles
205 increased the hydrophilicity, fouling resistance and mechanical strength of the CTA
206 membranes and improved the water flux by 65% (Chiao et al., 2019). In summary, compared
207 to commercial CTA membranes, membranes modified with nanoparticles give better results.
208 However, care should be taken to decide the type and the concentration of nanoparticles for
209 optimized FO performance, because some studies have shown that overloading carbon
210 nanotubes on the FO membrane had a negative impact on performance.

211 ### 4.5. Modifications to TFC membranes

212 Researchers have also modified the active and support layers of the TFC membrane for
213 similar reasons as with the CTA membrane (Section 4.3). However, modified TFC membranes
214 gave better mechanical and chemical stability, prompting a lot of research into TFC membrane
215 modification (Suwaileh et al., 2020).

216 Over the years, there has been much research to make the active layer more selective,
217 hydrophilicity and resistant to fouling (Shakeri et al., 2019b). Organic chemicals like
218 polyethylene glycol block polymer (Xu et al., 2019), phenylenediamine and trimethyl chloride
219 (Zhou et al., 2020) were successfully used to improve the hydrophilicity of the membrane. The
220 active layer of TFC membrane modified with these organic polymers (Fig. 2) also improved

the fouling resistance and structural parameters, which in turn yielded higher water flux (Xu et al., 2019).

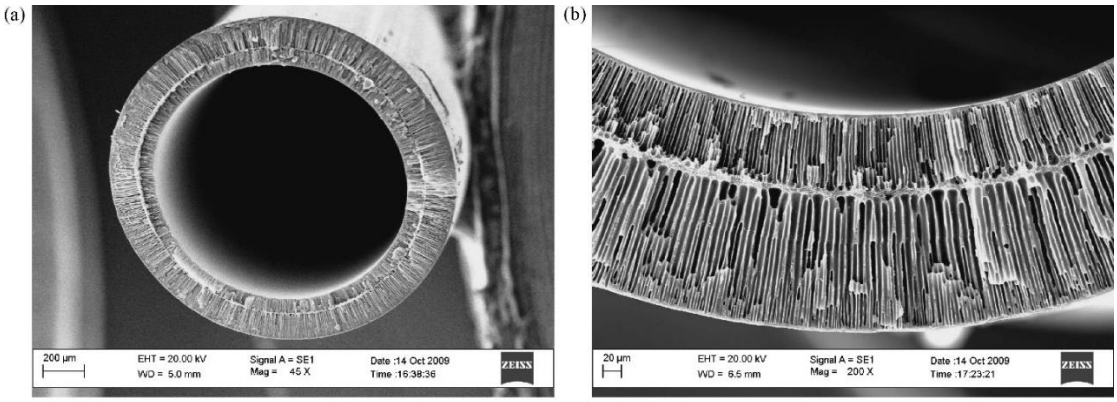

Fig. 2. Image depicting the hollow polymer fiber membrane used in FO (Wang et al., 2010)

Modifying the active layer surface by a novel composite of N-[3-(trimethoxysilyl) propyl] ethylenediamine and m-phenylene also increased water flux due to improved hydrophilicity and antifouling properties (Zhou et al., 2020). Coating by monomers like Zwitterionic amides was found to increase selectivity (Zhang et al., 2016) and enhance the anti-fouling and anti-microbial efficiency in desalination of industrial greywater (Zhang et al., 2017). It also caused a reduction in structural parameters which enhanced water transport characteristics (Khorshidi et al., 2016).

Motivated by the benefits of the active layer modification, researchers modified the support layer of the TFC membrane to improve the stability, porosity, durability, tortuosity, pore size and strength of the membrane (Xu et al., 2019). Polyacrylonitrile with fine pore size, used in the support structures of the TFC membrane, enhanced the water flux due to its improved porosity and hydrophilicity (Chiao et al., 2019). Similarly, ester (Zhou and Lee, 2016), polyketone (Yasukawa et al., 2017) and acetone (Zhang et al., 2016) doped in the support layer of the TFC membrane generated high water flux by reducing the structural parameter and improving the pore size, porosity and tortuosity (Rastgar et al., 2017).

Sulfonated polymers have also been used to improve the mechanical strength in the support layer of the TFC membrane (Qiu et al., 2011). For example, di sulfonated poly arylene ether sulfone and polyethersulfone improved the tensile strength and porosity in the support layer of the membrane increasing the water flux 16.4 LMH of 40 LMH (Lotfi et al., 2015). Another modification in the support layer is the use of double or multi-skin membranes over the support layer, which reduces the fouling and protects the inner membrane from interacting with DS (Ren and McCutcheon, 2015). For example, a double-layer membrane system with polyvinylidene fluoride and polyvinyl chloride produced a water flux of 65 LMH and 55 LMH, respectively (Tian et al., 2017). Future research should optimize the concentration of the polymers and monomers used in the active and support layers of the TFC membrane.

4.6.Addition of nanoparticles to TFC membrane

Nanoparticles have been coated on TFC membranes to improve surface morphology, membrane stability, water permeability, and hydrophilicity (Liang et al., 2017). For example, titanium and titanite nanotubes fabricated on the polyamide active layer of TFC membranes enhanced the surface morphology and hydrophilicity. This in turn increased the water flux and reduced reverse salt flux (Liang et al., 2017). The same authors found that titanium oxide nanoparticles enhanced the anti-fouling behaviour, selectivity and rejection capacity of the virgin TFC membrane

Carbon-based nanoparticles including graphene oxide (GO) and CNT improved the porosity, hydrophilicity and selectivity of the TFC membranes (Shokrollahzadeh and Tajik, 2018). For example, GO nanosheets in the polyamide active layer gave a thinner, smoother, and more hydrophilic selective layer with better structural parameters and improved water permeability (HG et al., 2017). Various nanoparticles have also been added to GO to improve further the membrane properties (Faria et al., 2017). For example, a blended composite of both CNT and graphene oxide on the active layer formed wide finger structures in the pores, due to

265 which, the surface properties like hydrophilicity and membrane selectivity were significantly
266 improved (Shokrollahzadeh and Tajik, 2018). This allowed a high water flux of 139 LMH.
267 Polyvinyl pyrrolidone was further used to modify graphene oxide-coated membranes as it
268 reduces GO aggregation and helps the homogenous distribution of GO on the TFC membrane.
269 Silver oxide nanoparticles with GO improved the anti-biofouling property of the TFC
270 membrane, which in turn increased the water flux by 80% (Faria et al., 2017).

271 For prolonged usage or continues operation of the TFC membrane in practical
272 applications, polyacrylonitrile (Pan et al., 2017) nanofibers were used in the active layer of the
273 TFC membrane. These nanofibers made the selective layer more hydrophilic and enhanced its
274 wettability, which prevented the TFC membrane from swelling during prolonged use (Pan et
275 al., 2017). With the addition of polyacrylonitrile nanofibers, the water flux was enhanced due
276 to its reduced porosity and structural parameters (Tian et al., 2017).

277 Besides coating the active layer, researchers have also tried to coat nanoparticles on the
278 support layer of the TFC membrane to analyse the variations in properties like surface area,
279 stability, porosity, durability, tortuosity, pore size and strength of the membrane (Rastgar et al.,
280 2017). Because of the high hydrophilic nature and stability of GO, CNT and silica nano
281 substrates, these nanoparticles have also been used by various researchers to modify the support
282 layer of the TFC membrane (Park et al., 2015). GO reduced the structural parameter from 217
283 µm to 163 µm and provided better thickness and porosity. GO-coated support layer TFC
284 membrane generated a high-water flux of 41 LMH (Wang et al., 2015). CNT and halloysite
285 nanotube were used to reduce the fouling by 60-70% in the support layer, thus enhancing the
286 water flux (Zhang et al., 2016).

287 Nanoparticles like calcium carbonate (Kuang et al., 2016), zinc (Zhao et al., 2017) and
288 silver oxide nanoparticles are more available commercially than GO and CNT. They have been
289 used in the support layer (Qiu and He, 2018) to improve the hydrophilic nature and structural

parameters of the membranes (Qiu and He, 2018). Recently, silica nanoparticles i.e. mesoporous materials (Fig. 3) were embedded in the nanofiber support layer of the TFC membrane increased the tortuosity, porosity of the virgin TFC membrane (Shakeri et al., 2019b). , which in turn resulted in a high water flux of 72 LMH (Lee et al., 2015).

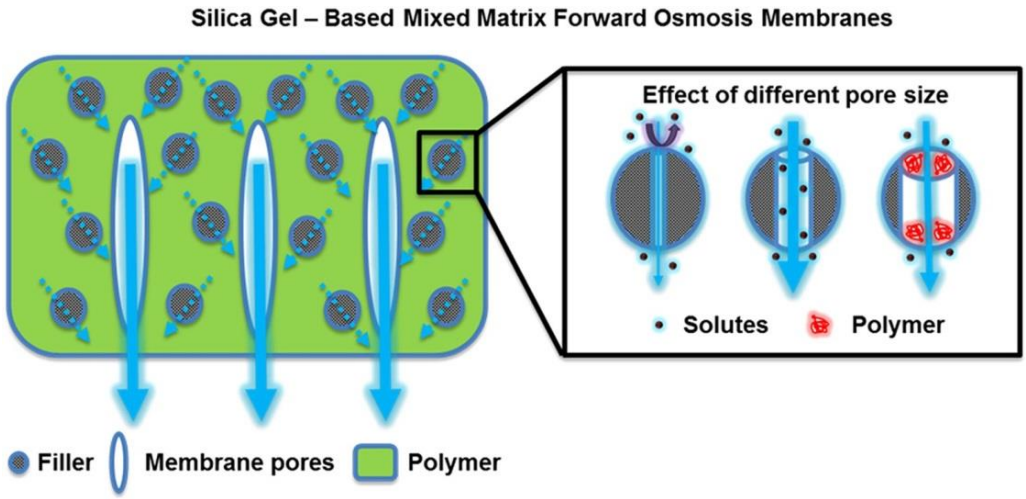

Fig. 3. Mesoporous silica membrane in FO system (Lee et al., 2015)

Layer-by-layer support (e.g. double- or triple-layer) has also been used to improve the properties of the TFC membrane (Xu et al., 2015). For example, hydroxide nanoparticles in double-layered created microvoids and formed finger-like structures in the TFC membrane, which improved water permeability and reduced the structural parameter (Duong et al., 2013). In another instance, three-layer support was provided by chitosan, polyacrylic and polyelectrolytes on the support layer of the TFC membrane improved the morphology, hydrophilicity and selectivity of the virgin TFC membrane, which resulted in a high-water flux (Pardeshi and Mungray, 2014).

Whereas most modifications were applied to either the support or active layer individually, in some studies aluminium oxide nanoparticles were applied to both layers of the TFC membrane (Ding et al., 2017)., thus improving the surface morphology, pore size, porosity, fouling resistance, high roughness and permeability (Kotp, 2021). In summary, modified TFC

membranes showed better performance with improved membrane properties and surface morphology, making them well suited to continuous operation. Future research should be directed towards optimising the volume and weight fractions of individual nanoparticles for each application.

4.7. Membrane orientation

Membrane orientation is one of the major parameters that govern the water flux of the FO process. The two possible orientations are (1) active layer of the membrane facing feed solution (AL-FS) (Fig. 4a) and (2) active layer facing the draw solution (AL-DS) (Fig. 4b) (Pramanik et al., 2019). AL-FS membrane orientation is preferred when treating hypersaline water, including wastewater from textile industries (Korenak et al., 2019), radioactive waste (Liu and Wang, 2013), pharmaceutical wastewater (Dong and Ge, 2019) and seawater (Valladares Linares et al., 2014). In contrast, AL-DS membrane orientation is preferred in treatment of low salinity water like brackish water (Zhao et al., 2012). Since AL-DS membrane orientation is preferred treating low salinity water, the quality of desalinated permeate was increased.

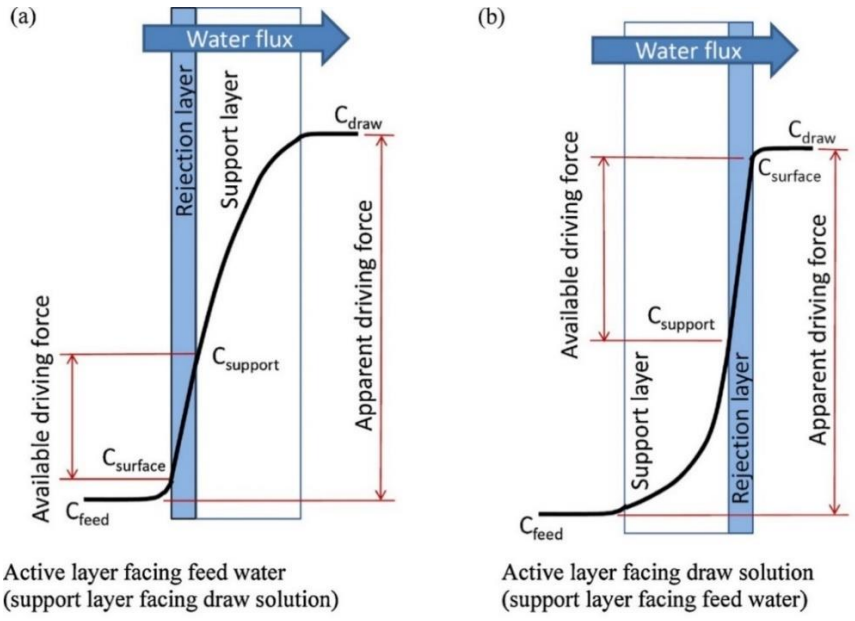

Fig. 4. Schematic of membrane orientation (a) AL-FS and (b) AL-DS (Xu et al., 2010)

AL-FS oriented membrane has shown better ability to maintain water flux in applications prone to fouling and scaling (Hawari et al., 2016). To summarize, concerning membrane orientation, AL-FS membrane orientation is better for treating high saline water and enhancing the water flux; whereas AL-DS membrane orientation is better to treat low saline waters like brackish water.

4.8.Limitations of FO membranes

Fouling and concentration polarization (Zhang et al., 2012) are two major limitations that affect the membrane performance and reduce water flux (Tripathi, 2015). Researchers have therefore sought to investigate and mitigate these phenomena, as discussed next.

4.8.1. Fouling and fouling mitigation

Deposition of various contaminants on the surface of the FO membranes may result in fouling and loss of performance (Fig. 5) (Hizam et al., 2020). The main types of fouling are particulate (suspended or colloidal), scaling (Mi and Elimelech, 2010), organic and biofouling (Fortunato, 2020). For example, deposition of inorganic contaminants (particulates) like calcium and silica on the membrane surface contributes to inorganic fouling (Parida and Ng, 2013); whereas the deposition of alginate, albumin (Aftab et al., 2020), silica (W. J. Lee et al., 2020), humic acid and lysozyme (Yangshuo et al., 2013) contributes to organic fouling (Lee et al., 2010a); and the accumulation of bacterial contaminants like Pseudomonas aeruginosa on the membrane surface contributes to biofouling (Sim et al., 2018).

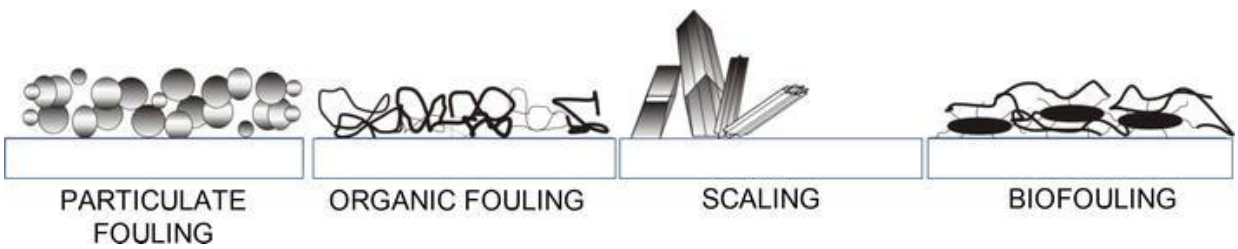

Fig. 5. Main types of membrane fouling (Fortunato, 2020)

To reduce or prevent fouling, operating parameters may be controlled, including hydrodynamic conditions (both DS and FS), temperature (both DS and FS) (Zou et al., 2013)., membrane orientation (Xie et al., 2013). For example, regarding hydrodynamic conditions (eg. flow velocity), increasing the DS flow rate from 5 L/h to 10 L/h decreased the water flux by 2% (C. Lee et al., 2020). Regarding temperature, at elevated temperatures (above 40℃), fouling is higher on the draw side as compared to the feed side (Fig. 6 a, b, c and d) due to higher permeability (Li et al., 2018) and more rapid diffusion on the feed side (Kim et al., 2015). Thus, a higher temperature may provide higher initial flux but, after a few hours, membrane fouling reduces the efficiency of the system (Zhao and Zou, 2011). So, temperature has to be optimised over a period of time. Regarding membrane orientation: AL-FS orientation gave higher water flux and tended to curtail fouling as compared to AL-DS orientation (Zhang et al., 2012). This was because the hydraulic pressure was more on the active layer than the support layer when the membrane faces the feed solution (AL-FS), which in turn decreases the deposition of foulant on the surface of the membrane (Zhang et al., 2012). In summary, maintaining the optimum temperature at both FS and DS and orienting the membrane in AL-FS orientation reduces the fouling (Boo et al., 2012).

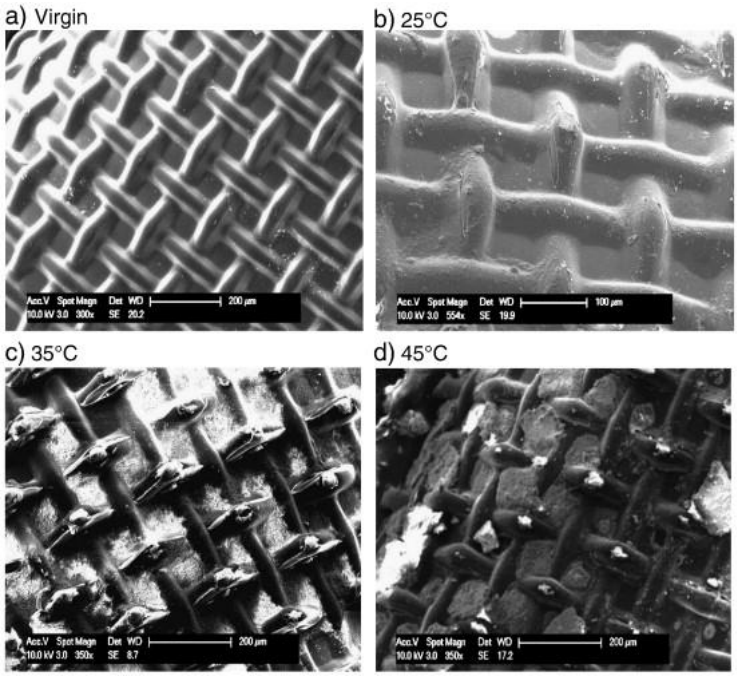

Fig. 6. Images depicting normal (a) and fouled (b, c, d) FO membranes from experiments conducted at various temperatures 25℃, 35℃ and 45 °C (Zhao and Zou, 2011)

Modifying the membrane surface is another approach to mitigate fouling (Emadzadeh et al., 2014). This includes changing the roughness, hydrophilicity and characteristic group of the FO membrane (Arkhangelsky et al., 2012). Reducing the roughness makes the membrane smoother, which makes it more difficult for the foulant to stick to the surface membrane (Arkhangelsky et al., 2012). Hydrophilicity can be increased by adding hydrophilic functional group materials, like grafting polyamine and ionic groups onto the membrane surface (Valladares Linares et al., 2011). These materials include aliphatic amine, mesoporous silica (Ramezani Darabi et al., 2018), carboxylate ions, iron oxide-zinc oxide nanocomposites and ethylene glycol (Y. Wang et al., 2016). Modifying the membrane through a hydrophilic functional group reduces the contact angle and increases the antifouling capacity (Choi et al., 2015).

Sometimes fouling is unavoidable, in which case periodic rinsing and cleaning may be used to reverse the fouling (J et al., 2020). As the fouling in FO is less dense as compared to that in

RO process, around 80-95% of the water flux can be recovered through proper periodic rinsing and cleaning (Lee et al., 2010b). Cleaning can be done either by tangential flushing of the membrane surface or by backwashing (W. J. Lee et al., 2020). Tangential flushing increases the shear force and cleans the foulant from the surface of the membrane (i.e. active layer). If the foulant is deposited in the support layer, then flushing and cleaning are not effective, and chemical treatment is preferred (Tow et al., 2018). Chemical agents like sodium hydroxide and nitric acid can be used to treat the membrane and to subsequently weaken the adhesive bond between foulant and membrane thereby mitigating fouling (Madaeni and Mansourpanah, 2004).

4.8.2. Internal and external concentration polarization

Concentration polarization refers to the accumulation of solutes near the membrane surface (Arjmandi et al., 2020). It slows down the FO process by decreasing the osmotic pressure and thus reduces the efficiency of the system (Wong et al., 2012). Whereas RO is only affected by external concentration polarization (ECP), FO is affected by both external and internal concentration polarization (ICP) (J. Wang et al., 2016). ICP occurs in the internal surface of the membrane i.e. the support layer, whereas ECP occurs in the external surface of the membrane i.e. in the active layer (Xuan Zhang et al., 2018). Membrane orientation affects the relative accumulation in the support and active layer, which gives rise to either concentrative or dilutive ICP and ECP (Gray et al., 2006) (Fig. 7). For example, if the membrane is in AL-FS orientation, then the concentration of DS becomes higher near the external surface (active layer) while the FS becomes less concentrated near the internal surface, contributing to concentrative ECP and dilutive ICP, respectively (Bhinder et al., 2018). Conversely, for a membrane with AL-DS orientation, concentrative ICP and dilutive ECP occurs (Abdelrasoul et al., 2018).

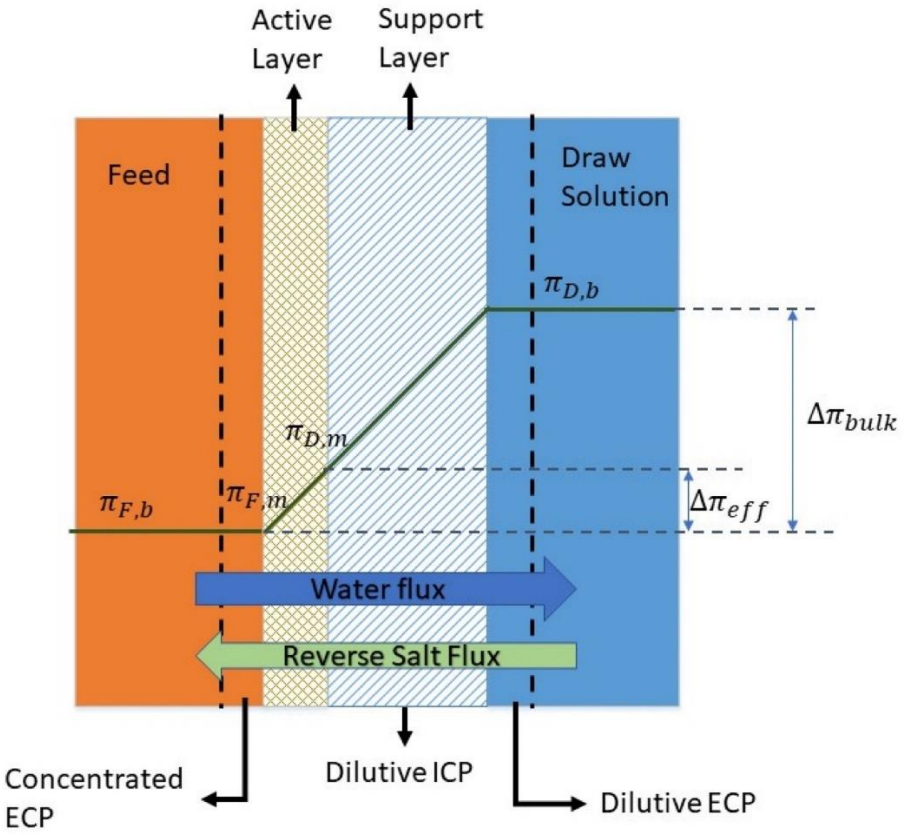

Fig. 7. External and internal concentration polarization developed in an FO membrane oriented in FO mode i.e. active layer facing the FS (Anjum et al., 2021).

ICP affects water flux more severely than ECP. For example, ECP (both dilutive and concentrative ECP) decreases the difference in DS and FS transmembrane pressure, which reduces the water flux only slightly (Bhinder et al., 2018). Whereas ICP (both dilutive and concentrative ICP) impacts the concentration of FS or DS (based on membrane orientation) and reduces the concentration difference between DS and FS affecting the water flux in greater proportion (Tang et al., 2010). Thus, reducing ICP and ECP is more important to achieve a greater water flux. In this section, we discuss research advances aimed at overcoming the problem of concentration polarization (ICP and ECP).

4.8.3. Overcoming ICP and ECP

Methods of mitigating ECP tend to aggravate ICP and vice-versa. As ECP occurs on the external surface i.e. active layer of the membrane, this can be controlled by increasing the flow

rate of the FS and DS (W. J. Lee et al., 2020). For example, Suh and Lee (2013) found that restricting the flow of DS into the membrane support layer reduced ECP, but also worsened ICP in the supporting layer (Suh and Lee, 2013). Increasing the flow rate and optimizing the flow rate equalizes the distribution of concentration across the membrane surface thus reducing ECP (Gruber et al., 2011).

As ICP occurs in the internal surface of the membrane (i.e. in the support layer), modification of the support layer such as a structural layer, temperature, functional group, tortuosity, wall thickness and porosity reduces the ICP in the FO membrane (W. J. Lee et al., 2020). For example, decreasing the wall thickness in the support layer and increasing the tortuosity reduces ICP (Tan and Ng, 2008).

In another instance, increasing the temperature difference across the membrane enhanced the water flux but, beyond a threshold of 40°C difference, the concentration polarization increased which in turn reduced the efficiency of the process (McCutcheon and Elimelech, 2006). Thus, maintaining an optimum temperature across the membrane decreases the ICP effects and increases the water flux (McCutcheon and Elimelech, 2006). This is because increasing the temperature helps to reduce the deposition of compact crystals on the membrane surface, which in turn increases the water flux. In summary, AL-FS membrane orientation with a temperature less than 40°C is usually preferred in FO to reduce ICP and to enhance the water flux (Tang et al., 2010).

5. Draw solution

The draw solution is one of the main components in FO, as it provides the driving force for the process (Qasim et al., 2017). High osmotic pressure, high solubility, and low viscosity are important criteria in DS selection (Jingxi et al., 2019). Almost all draw solutions are aqueous (Chaoui et al., 2019); therefore, the DS may be distinguished according to the solute

used. A few researchers have used volatile solutes such as NH_4CO_3 and SO_2 and dimethyl ether (Sato et al., 2014). However, due to their drawbacks like low water flux, high reverse solute flux and biofouling, most researchers have opted for non-volatile solutes that may be inorganic or organic type (McCutcheon et al., 2006).

5.1.Comparison of inorganic and organic DS

Inorganic DS are usually monovalent or multivalent ions that have high water solubility and are thus able to create high osmotic pressure (Jingxi et al., 2019). They include Mg^{2+}, Ca^{2+}, Ba^{2+}, SO_4^{-2}, KCl, $NaNO_3$ and NaCl (Johnson et al., 2018). Many standard solutes including Mg^{2+}, Ca^{2+}, Ba^{2+}, SO_4^{-2}, $NaNO_3$, $MgSO_4$, Ca $(NO_3)_2$, Al_2SO_4 and $CuSO_4$ are used in industrial processes but have limitations of scaling, fouling and high reverse solute flux (Shon et al., 2015). Specifically, Mg^{2+}, Ca^{2+}, Ba^{2+} and SO_4^{-2} groups cause membrane scaling and fouling which in turn reduces the water flux (Chekli et al., 2012). $NaNO_3$ had disadvantages of low solubility and high reverse solute diffusion (which contaminates the feed) (Nguyen et al., 2015). Other DSs such as $MgSO_4$, Al_2SO_4 and $CuSO_4$ were assessed and found to have poor performance with cost penalty and gave toxic by-products in the permeate (Ge et al., 2013). DS like $Ca(NO_3)_2$ and oxalic acid were found to have high biofouling properties and metal toxicity (Johnson et al., 2018). Among various inorganic DS, compared to multivalent salts, monovalent salts have high energy consumption for regeneration and possess high reverse solute flux due to their lower hydrated radius and electrostatic repulsion (Alejo et al., 2017). Because of this, researchers preferred multivalent salts as DS in recent years, including $MgSO_4$. $CaCl_2$ and fertilizers (e.g. $NH_4H_2PO_4$, and $(NH_4)_2SO_4$) (Alejo et al., 2017). Fertilizer used as DS gets diluted and can be mixed with water and used for irrigation (Phuntsho et al., 2011).

Organic DS include ionic liquid co-polymer (salts in liquid state). These have high osmotic pressure (Chen et al., 2019). The use of ionic liquids as draw solutions is helped by its hydrophilic properties (Dutta and Nath, 2018). Hydrophilic anions enhanced the osmolality at

elevated temperatures, which in turn increased the water flux (Kamio et al., 2019). Another ionic liquid, lightweight oligomeric co-polymer like poly-tetra butyl phosphonium styrene sulfonate was also used as DS to improve the DS recovery rate. This could be separated by just raising the temperature without the use of any other separation process (Kim et al., 2016). Around 99.5% water was recovered which makes this polymer a good candidate in organic DS in FO (Zhao et al., 2014). Another novel DS was made with co-polymer i.e. zwitterion group of poly-sulfobetaine, through free-radical polymerization (Pejman et al., 2020b). This novel draw solution overcame drawbacks related to reverse solute and low water flux (Pejman et al., 2020a).

5.2.Use of nanoparticles in DS

Besides the use of ionic liquids, researchers have also used functionalized nanoparticles to increase the water flux of DS (Na et al., 2014). For example, carbon nanofibers (Fig. 8) enhanced the water flux due to the higher osmotic pressure (Tavakol et al., 2020). Standard organic DS and DS based on functionalized nanoparticles are easy to regenerate and gave a good performance (Ling and Chung, 2011), but in some cases their toxic nature hinders their wider application, prompting researchers to investigate the use of hydrogels such as novel polymeric and copolymeric polymers (Dabaghian and Rahimpour, 2015).

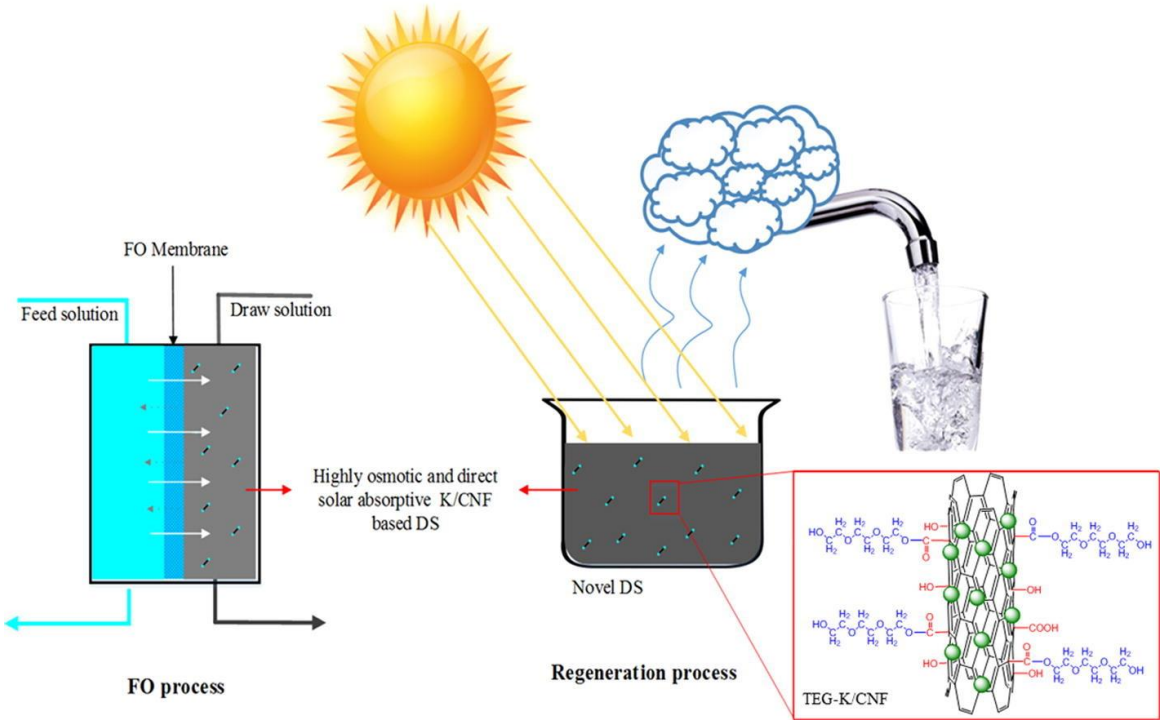

Fig. 8. A novel draw solution (tri-ethylene glycol suspended in potassium functionalized carbon nanofibers) for FO (Amjad et al., 2018)

5.3.Hydrogel-based DS

Compared to other organic DSs like diethyl ether and poly-tetra butyl phosphonium styrene sulfonate, hydrogels yield better water flux and give lower reverse solute flux with reduced concentration polarization (Hsu et al., 2019). For example, researchers have manufactured thermo-responsive hydrogels by polymerization of N-isopropyl acrylamide with poly sodium acrylate and used it as a DS in FO (Fig. 9a and b). The resulting hydrogel had a superior swelling and dewatering behaviour (Wibisono and Bilad, 2019). These types of thermo-responsive hydrogel (Fig. 9c, d and e) consume less energy and are recommended for industrial applications (Cai et al., 2013). Some researchers have manufactured hydrogels by ring-opening polymerization involving monomers of triblock copolymer including graphene oxide. The results showed that hydrogel having 0.09% weight graphene oxide gave the best results when used as DS (Nakka and Mungray, 2016). The research concluded that graphene in the hydrogel

could increase the swelling ratios and water fluxes due to the presence of functional groups that enhance the hydrophilic nature of the modified polymer hydrogels (Nakka and Mungray, 2016).

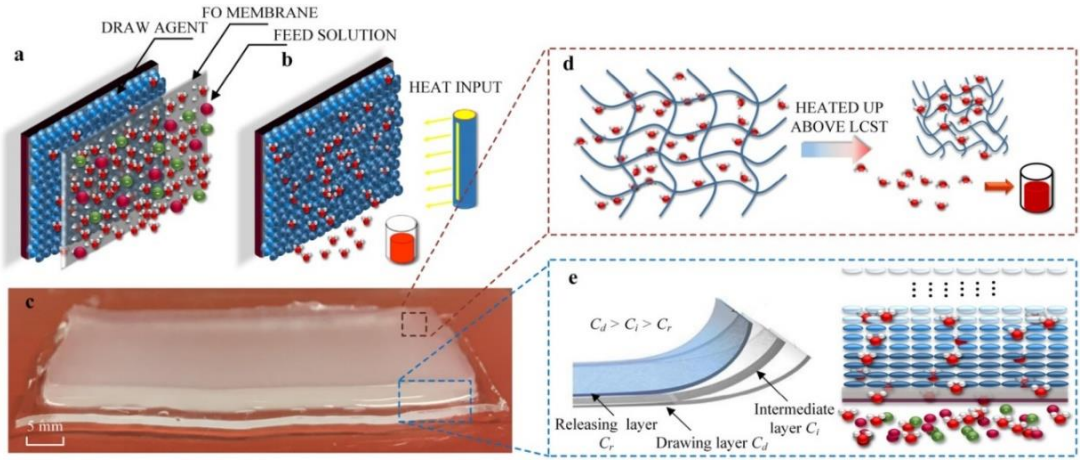

Fig. 9. Schematic of (a) FO membrane (b) water releasing with temperature input (c) novel hydrogel (d and e) multi-layer design with water transportation (Zeng et al., 2019)

Motivated by the benefits of hydrogel, researchers compared four new hydrogel polymers (two ionic, two non-ionic) with respect to water flux. Two ionic hydrogels (poly-sodium acrylate and N-isopropyl acrylamide) and two non-ionic hydrogels (acrylamide and N-isopropyl acrylamide) were investigated (Li et al., 2011). N-isopropyl acrylamide gave the highest flux because of its high dewatering rate (Li et al., 2011). Building on the research work of Li et al., (2011), researchers tried to enhance hydrogels with graphene oxide and optimized the weight percentage of graphene oxide (Zeng et al., 2013). The results showed that 1.2% of reduced graphene oxide in hydrogel could increase the swelling ratio, which in turn increased the water flux. Another reason for such enhancement was increased softness and inter-particle and particle-membrane contact of the composite hydrogels, which led to an enhanced water flux at the output. In addition, the thermal properties of graphene oxide, especially thermal conductivity, helped to dewater the hydrogel giving an increased water recovery rate (Zeng et al., 2013).

Another attempt was made with copolymer i.e. sodium styrene-4-sulfonate-co-n-isopropyl acrylamide (with 15 weight percentage) to get the maximum flux (Zhao et al., 2014). The reason for this enhancement was that, as the temperature increased, the water vapour pressure also increased whereas the osmotic pressure decreased, which enabled an effective regeneration of water in the membrane and consequently enhanced the water flux further (Zhao et al., 2014).

In summary, many novel hydrogels have been developed to replace the conventional DS and to reduce concentration polarization thus improving water recovery. Poly-sodium acrylate and N-isopropyl acrylamide are especially promising, as they achieved a very high water flux of 74.2 LMH (Kamio et al., 2019). Moreover, other operational parameters like concentration, temperature, and cross-flow velocities also have an important effect when using these novel DS.

6. FO operating parameters

The performance of FO significantly depends on various operating parameters which include operating temperature, flowrate and concentration (Wang and Liu, 2021). Studies have been conducted to optimize these parameters with respect to both FS and DS, as discussed in the next section:

6.1.What operating temperature range is best for FS and DS?

Several studies, both experimental and theoretical, have been carried out to optimize the operating temperature of DS and FS for enhanced output (Fig. 10).

Experimental results have shown that water flux increases with temperature but reverse solute flux may also increase (Traxler, 1928). For example, several studies with NaCl as DS gave a 100% increase in flux on increasing the DS temperature from 5°C to 25°C (Adhikary et al., 2020). A different study with NaCl obtained only a 21% increase when the DS temperature

was increased from 25°C to 45°C (Phuntsho et al., 2012). The flux increased by an even smaller 15% when a similar temperature increase was applied on the FS side (Arcanjo et al., 2020). The reason for the smaller flux enhancements could be the reverse solute flux which increased by 47.5% with NaCl forward rejection of 1.2% under temperature increase on the DS side, compared to just 8.3% and 0.4%, respectively, when temperature was increased on the FS side (Phuntsho et al., 2012).

Another study found that water flux was increased by 100%, on increasing both DS and FS temperature from 3℃ to 40℃, because of the enhanced mass transfer across the membrane (McCutcheon and Elimelech, 2006). Increasing the temperature of FS and DS above 40°C further improved the water flux in FO due to the enhanced diffusion kinetics. The viscosity decreases with temperature, which in turn increases the water permeability across the membrane (Wang et al., 2014). Thus, higher temperatures may provide higher initial flux; however, after a few hours, membrane scaling and fouling reduces the efficiency of the system (Zhao and Zou, 2011). Another contradictory finding is that the FO system (both FS and DS) which operated at 26℃ gave only a 36% enhancement of water flux compared to operation at 23℃, whereas just by decreasing the temperature to 23℃ (with all the other parameters similar), there was a significant enhancement of 72% (Hawari et al., 2016). The difference of only 3℃ in the operating temperature apparently caused a large enhancement in the water flux – a finding that may justify further investigation (Xie et al., 2013). Various DSs including KCl, NaCl, $CaCl_2$ and Na_2SO_4 have been experimentally tested in FO to determine water flux over a range of temperature from 25 to 40°C (Heo et al., 2016). KCl and NaCl gave higher water flux than other DS, because of the hydration radii of chloride and sodium ions (C. Wang et al., 2019).

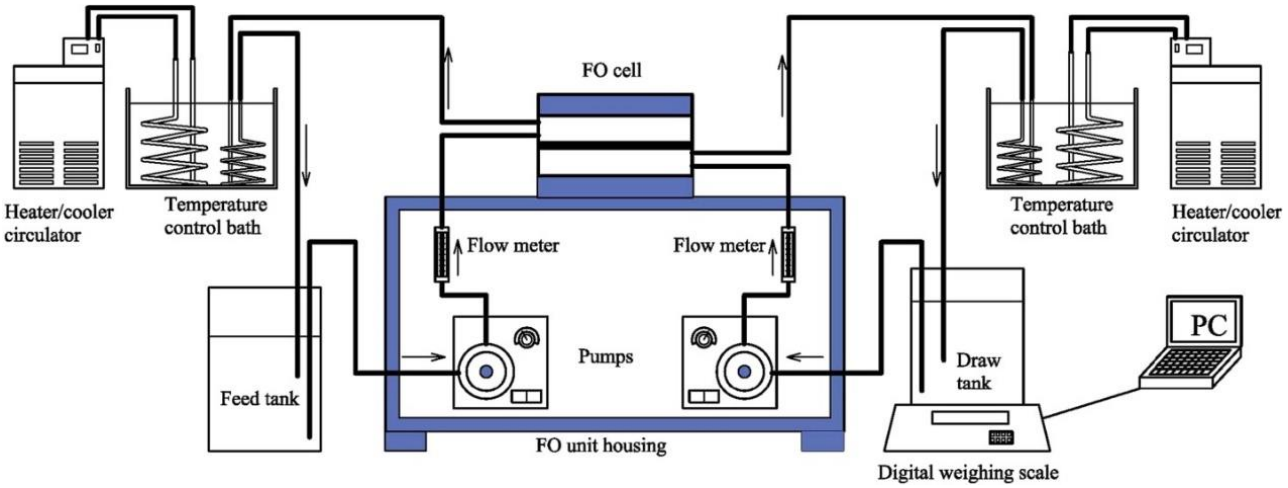

Fig. 10. Schematic of temperature-controlled FO setup (Phuntsho et al., 2012)

Besides carrying out experimental studies, researchers have used model-based prediction to investigate the effect of temperature on water flux and reverse solute flux (Akbari and Peyravi, 2020). The results predicted that the viscosity of the solution is inversely proportional to the temperature; whereas the diffusivity is directly proportional to the coefficients of pure water and solute permeability (Chowdhury and McCutcheon, 2018). The accuracy of the model was further improved by including the effect of heat transfer across the membrane (Jawad et al., 2021), external resistivity (boundary layer) on the active (Nagy, 2014) and support layer of the FO membrane (Nagy et al., 2014). The simulations showed that an increase in feed temperature near the membrane active layer from 20℃ to 40℃ was responsible for increasing the salt permeability (by around 150%) and water permeability (by around 161%) across the membrane (Lee and Ghaffour, 2019). To analyse the effect of temperature more precisely, a steady-state model was used to measure the temperature variation concerning the changes in osmotic pressure and water dynamics of the boundary layer in the DS (You et al., 2012). Since the developed steady-state model considers multiple parameters together like hydrodynamics of the boundary layer, temperature variation on osmotic pressure, mass transfer across the membrane and heat flux across the membrane to predict the water flux of the system, it is likely to be very precise (Ettouney and Aldaihani, 2020),. The model showed that an

increase in temperature reduced the viscosity and accelerated diffusion kinetics across the membrane, which in turn increased the water flux and performance of the process. It also showed that a decrease in the transmembrane temperature caused a decrease in the water flux due to the reduction in transport kinetics and osmotic pressure (You et al., 2012).

In summary, increasing the temperature (DS or FS side) increases the water flux and decreases the reverse solute flux. This is because increasing the temperature of DS or FS decreases the viscosity of the fluid, causing the diffusion of the molecules to increase, which in turn increases the water flux. The temperature may be beneficially increased up to 40°C (depending on details such as DS concentration and membrane properties) to reduce concentration polarization in the FO system.

6.2.Optimal flowrate

Flow rate of FS and DS influence various operational parameters like concentration polarization, water flux and efficiency (Heo et al., 2013). Higher flow rates reduce the concentration polarization, which in turn impacts the water flux and recovery of the FO system (Ahmed et al., 2019). This variation in water flux depends on the orientation of the membrane (i.e. either AL-DS or AL-FS). Increasing the flow rate at the draw side alone (from 1.2 L/min to 3.2 L/min) reduces the water flux by 52% if the membrane is in AL-FS orientation. This reduction may be attributed to the increased turbulence on the support layer of the membrane and back diffusion of the salt (Tow et al., 2018). Whereas with AL-DS orientation, increasing the flow rate (from 1.2 L/min to 3.2 L/min) at the draw side alone enhances the water flux by 36% (Hawari et al., 2016). This enhancement may be due to the reduced external concentration polarization on the membrane surface (Mat Nawi et al., 2020). At the same time, with AL-DS orientation, increasing the flow rate at the feed side alone (from 1.2 L/min to 3.2 L/min) increases the flux by 38.5%. Here the enhancement may be due to reduced internal

concentration polarization and fouling in the support layer of the membrane (Mazlan et al., 2016).

Attempts were also made to investigate the effect of water flux on increasing the flow rate at both DS and FS simultaneously with AL-DS orientation. There was a 76% enhancement in the water flux achieved with the increase in the flow rate of both DS and FS simultaneously from 1.2 L/min to 3.2 L/min (Hawari et al., 2016). Even though increasing the flow rate may mitigate the concentration polarization and enhance the water flux, this may augment the energy consumption for the pumping process. Reducing the flow rate of DS from 100 mL/min to 10 mL/min decreased the energy consumption from 1.86 kWh/m^3 to 0.02 kWh/m^3 (Zou and He, 2016). In summary, increasing both DS and FS flow rate simultaneously was 40% more efficient than increasing either of the flow rates individually. This may be attributed to the simultaneous reduction of ICP and ECP.

6.3.Effect of varying FS and DS concentration

A difference must be maintained between concentration of FS and DS to maintain the osmotic pressure gradient which drives the high water flux (Cui et al., 2014). Hence research has been conducted to analyse the effect of varying the FS and DS concentrations on water flux (Li et al., 2013).

Water flux has both linear and non-linear relations with the DS concentration. The type of DS and its concentration are important factors to get higher water flux (Cornelissen et al., 2008). For example, increasing the concentration of some DSs like NaCl and MgCl showed a linear relation to water flux, whereas other DSs like $ZnSO_4$ and EDTA showed a non-linear relation (Cornelissen et al., 2008). When NaCl concentration increased from 0.5 to 4.5 M, the water flux increased from 6 LMH to 13 LMH (Hau et al., 2014). In comparison, when $ZnSO_4$ concentration increased by the same amount, the water flux increased from 2.2 LMH (at 0.5

M) to 4.5 LMH (at 2.3 M), beyond which the water flux remained unchanged even when the concentration was raised to 4.5 M of $ZnSO_4$ (Cornelissen et al., 2008). This difference may be due to ICP, which was higher for $ZnSO_4$ because of its lower diffusion coefficient. For EDTA as DS, the water flux increased from 4 LMH to 13 LMH when DS increased from 0.1 M to 1 M (Hau et al., 2014). The water flux varied linearly up to 0.7 M EDTA, beyond which the variation in water flux was increased in a negligible manner, which accounted for just a 3% increase in water flux from 0.7 M to 1 M of ETDA (Hau et al., 2014). However, for the DS which has higher diffusion coefficients, high reverse salt flux is one of the major limitations while increasing the DS concentration. Hence, selection of DS concentration should be done based on high diffusion coefficient and low reverse salt flux for getting higher water flux (Phuntsho et al., 2013). In summary, increasing DS concentration is preferred over increasing FS concentration and optimum concentration of DS depends on the type of DS selected.

7. FO applications and hybrid systems

Many different applications of FO have been researched in recent years, including use in multistage flash (MSF) desalination plants (Cath et al., 2006), pressure retarded osmosis (PRO) (Loeb, 1976), biomass industries (Alsvik and Hägg, 2013), food processing industries (Haupt and Lerch, 2018), and pharmaceutical industries (PRO) (Cui and Chung, 2018). In MSF desalination plants (Fig. 11), FO is used as a pre-treatment technique (Darwish et al., 2016) for effective treatment of highly saline water (Altaee et al., 2013). In PRO, brine is directed to a high-pressure chamber, separated from an adjacent feed chamber by a semi-permeable membrane, such that water at high pressure and high concentration draws water towards the high-pressure chamber thus performing mechanical work (Cheng et al., 2018). The feedwater can be river water or municipal wastewater effluent (Loeb et al., 1976),. A major disadvantage of PRO is its fouling propensity and FO can be used as a pre-treatment to alleviate this problem. The resulting hybrid FO-PRO system (Fig. 12) is less prone to fouling, and lower energy

requirements, thus increasing its overall efficiency several-fold (Jamil et al., 2016). Variants of FO, such as pressure-assisted FO (Fig. 13), have also contributed to enhanced efficiencies (Helfer et al., 2014). In biomass industries, FO can be used to treat the nutrient-rich wastewater extracted from the digested biomass (Holloway et al., 2007). Pharmaceutical industries also use FO, in this case, to concentrate the medical ingredients (Rastogi, 2019) and recover the organic solvent (Cui and Chung, 2018).

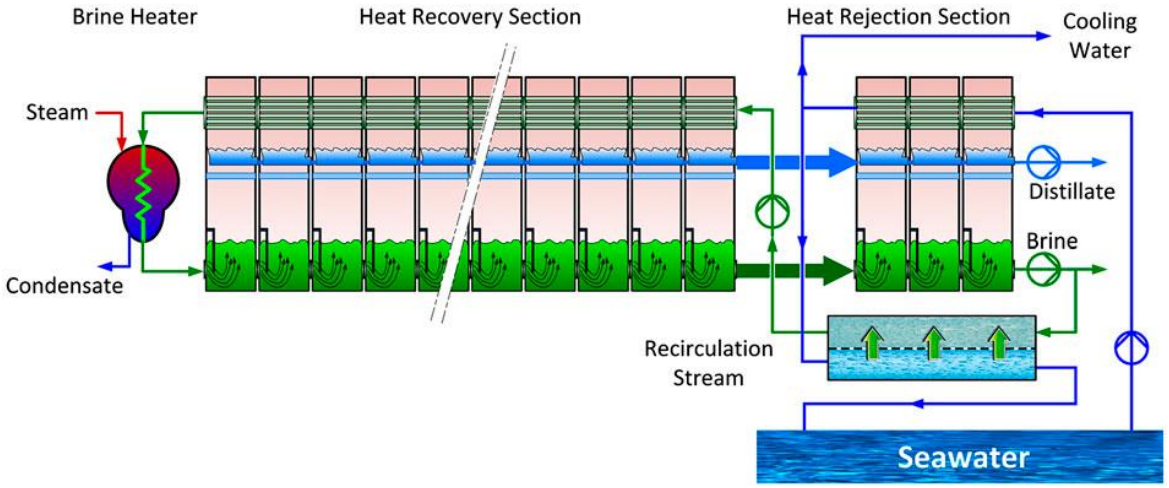

Fig. 11. Forward osmosis as a pre-treatment method for MSF system (Darwish et al., 2016)

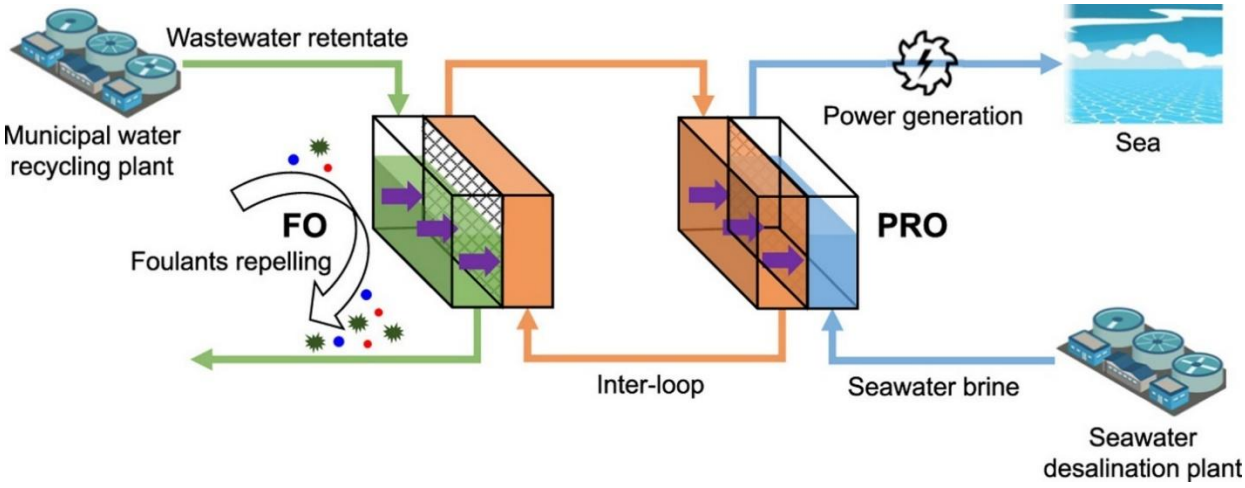

Fig. 12. Integrated FO-PRO system using wastewater retentate as FS and brine from seawater as DS for osmotic power generation (Cheng et al., 2018)

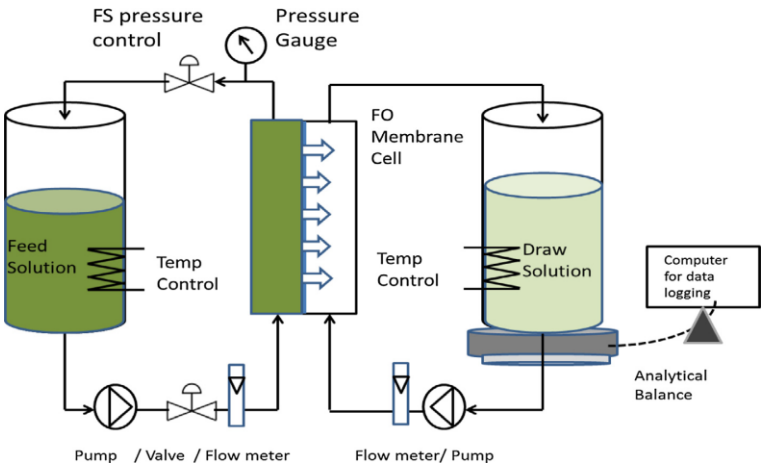

Fig. 13. Pressure assisted FO (Jamil et al., 2016)

FO has been integrated with other systems to form a hybrid system, to enhance efficiency (Usman et al., 2021) and to reduce the energy consumption of the whole system (Blanco Gálvez et al., 2009). For example, FO was integrated with membrane distillation units powered by solar energy (Fig. 14) (Q. Li et al., 2019). This integrated system reduced the specific energy consumption to 1.1 kWh/m^3 compared to 7.06 kWh/m^3 for the unmodified membrane distillation system (Suwaileh et al., 2019).

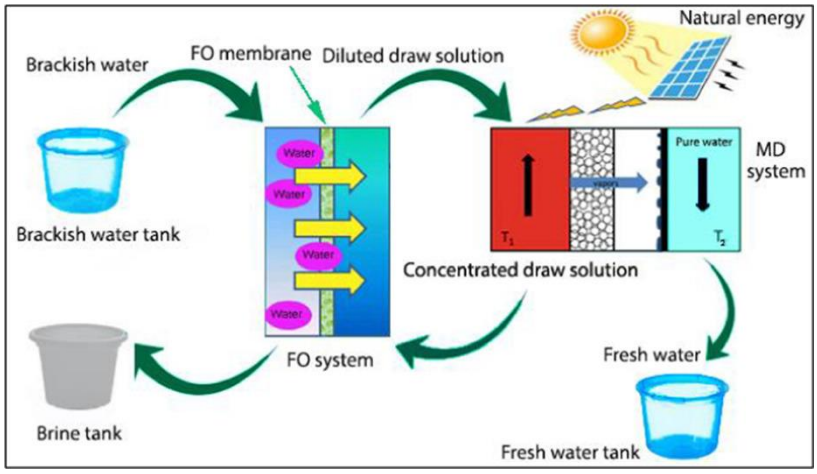

Fig. 14. FO powered from solar energy for reducing energy consumption (Suwaileh et al., 2019)

In another study, a multi-stage flash desalination unit was coupled with FO pre-treatment for seawater. The recovery of the hybrid system was enhanced by 32%; and

concentrations of calcium, magnesium and sulfate ions in the feed solution were reduced. Due to the lower concentration of these ions, water permeability and the water flux were enhanced in the hybrid system (Altaee et al., 2014a). Researchers have also coupled low-pressure reverse osmosis (RO) with a FO system to reduce the energy consumption from 2.5 kWh/m³ (for a standalone system) to 1.5 kWh/m³ (for the hybrid system). The fouling was reduced in the hybrid system thus yielding a greater water flux (Yangali-Quintanilla et al., 2011).

Studies reveal that, upon prolonged use, the decline of water flux in FO was less than in RO (Lee et al., 2010a). With sufficient DS concentration, the water flux in FO can even exceed that in RO (Altaee et al., 2014b). The FO membrane was more resistant to fouling than the RO membrane (Siddiqui et al., 2018), allowing high flux to be maintained over a longer period (Xie et al., 2017). Under standard conditions, RO gave better water flux as compared to FO but, on increasing the temperature and cross-flow velocities, the water flux for FO was several times higher than for RO (Altaee et al., 2017). This was attributed to increased fouling in RO at high temperatures (Mazlan et al., 2016). The RO system was coupled with FO to reduce the scaling, thus avoiding the use of chemicals in pretreatment and effectively remove the byproducts of sodium (such as hydrogen phosphate and hexametaphosphate) (Bamaga et al., 2011). The RO-FO system (Fig. 15) reduces the reverse water flux when operated at a higher temperature and the specific energy consumption of this hybrid system was only 1.66 kWh/m^3 – as such lower than conventional seawater RO which typically consumes >2 kWh/m^3 (Park et al., 2018). More than 60% of this energy was used for thermal heating of the flow stream (Ju and Kang, 2017) and hence the study recommended that the energy consumption could be further reduced by coupling FO with waste heat recovery devices (Wang et al., 2014).

Researchers have also integrated FO with biotechnological applications (Ma et al., 2020). For example, FO was used in a micro-algae dewatering system to achieve a water flux

of about 15.5 LMH with a recovery rate of 97% (Fig. 16). A stirrer was used to reduce the concentration polarization and enhance the water flux in this system (Zhang et al., 2019).

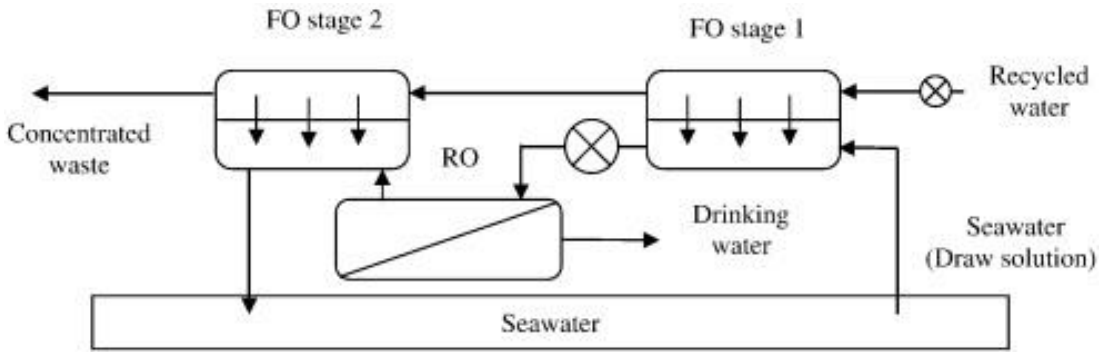

Fig. 15. Integrated system with FO and RO for augmenting the water flux and osmotic pressure (Bamaga et al., 2011)

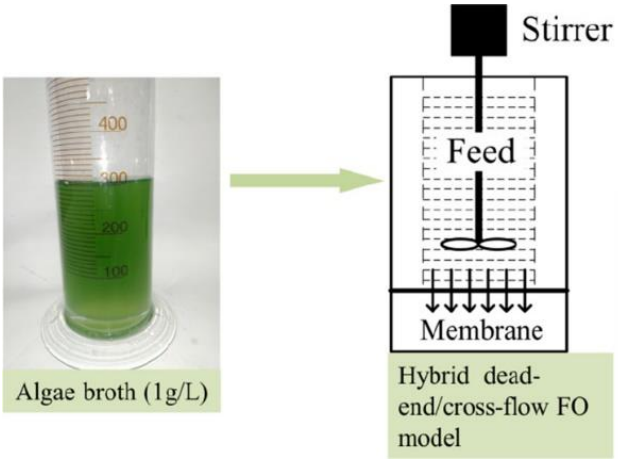

Fig. 16. A hybrid FO with micro-algae dewatering system (Ma et al., 2020)

In summary, FO can be integrated with various advanced technologies to improve their efficiency and reduce their energy consumption (Esmaeilion, 2020). The integration also reduces the concentration polarization with an improvement in the recovery rate of freshwater. In all cases, it is evident that hybrid systems are very advantageous over standalone systems. Further research should be conducted to use waste heat recovery devices for hybrid integration with FO, to enhance the performance. Table 1 gives an overview of studies concerning the working temperature, DS, type of membrane and water flux obtained.

717 Table. 1. Overview of forward osmosis desalination studies showing the working temperature, draw solution used, type of membrane and the flux

718 obtained (where data are provided).

Sl. No	Reference	Working temperature (highest) in ℃	Draw Solution	Advantages of the DS	Disadvantages of the DS	Type of FO membrane	FO membrane material	Water flux recorded (LMH)
1	(Phuntsho et al., 2012)	45	KCl	Enhanced osmotic pressure Low viscosity	Low water recovery	CA FO membrane	cellulose acetate	7.1
2	(Feng et al., 2018)	40	ammonium bicarbonate solution	Better water flux Decreased reverse solute flux	Enhanced ECP effects	asymmetric FO membrane	Asymmetric FO membrane, Saehan (Republic of Korea)	11.6
3	(Chowdhury and McCutcheon, 2018)	40	NaCl in DI water	Better mass transfer rates	Increase fouling after continuous use	CTA FO membrane	Cellulose triacetate	27
4	(Lee and Ghaffour, 2019)	40	NaCl	Decrease reverse salt flux Decreased fouling	High Cost Complex setup	TFC-PA FO membrane	Isopropyl alchol	27.26
5	(You et al., 2012)	40	NaCl	Increased transmembrane flux	Low working temperature range	CTA FO membrane	Polyester screen mesh with Cellulose triacetate	30
6	(C. Wang et al., 2019)	50	NaCl (1M)	High osmotic pressure	Low water recovery	CTA FO membrane	Cellulose triacetate	27.1
7	(Heo et al., 2016)	45	CaCl2	Low biofouling Low viscosity	Drastic decline in flux after continuous use	CTA FO membrane	Cellulose triacetate	24.9
8	(Li et al., 2018)	25	CaCl2	Enhanced water flux Low reverse solute flux	Low cross-flow velocity	TFC membrane	Cellulose triacetate. Active layer of polyamide and support layer of polysulfone	62
9	(Kim et al., 2015)	50	NaCl (3M)	Better diffusivity Enhanced Water recovery	Low durability	cellulose-based polymers membrane	Cellulose-polyester	29.61

10	(Xie et al., 2013)	40	NaCl (0.5M)	Reduced ICP effects	Increase desalination time	TFC membrane	Polyamide active layer with polysulfone support layer	27.3
11	(Zhao and Zou, 2011)	45	Na2SO4	Better water recovery	Increased fouling Not commercially available	CTA FO membrane	Cellulose triacetate	17 .
12	(Q. Wang et al., 2019)	47	NaCl (5M)	High water flux High osmotic pressure	Increased ICP effect	TFC membrane	-	71
13	(Ahmed et al., 2019)	85	ethylene oxide-propylene oxide copolymer	Fouling resistant	Not commercially available	CTA FO membrane	Cellulose triacetate	-
14	(Tow et al., 2015)	100	NaCl	Commercially available	-	-		-
15	(Xu et al., 2010)	24	NaCl	Low cost	low osmotic pressure	Commercial membrane	Cellulose triacetate	6.6
16	(Amjad et al., 2018)	50	TEG-K/CNF	High solar absorptive Better morphology	Complex setup	asymmetric FO membrane	-	13.3
17	(Wang et al., 2017)	45	Poly epoxy succinic acid	High rejection rate Reusability	High cost	TFC FO membrane	-	23
18	(Cai et al., 2013)	40	semi-IPN hydrogels	Low fouling reusability	Low water flux	CTA FO membrane	Cellulose triacetate	0.24
19	(Nakka and Mungray, 2016)	40	Triblock Copolymer Hydrogels	Biodegradable Anti-biofouling	Low water flux	CTA FO membrane	Cellulose triacetate	0.68
20	(Li et al., 2011)	50	Polymer hydrogel	High water flux Low reverse solute flux	Low durability	-		74.2
21	(Zeng et al., 2013)	45	polymer-graphene	Enhanced recovery	Membrane fouling	CTA FO membrane	Cellulose triacetate	8.2

			composite hydrogels	Low cost	Complex setup			
22	(Zheng et al., 2020)	45	Glycine	Enhanced water permeability High rejection rate	ICP effects	TFC FO membrane	Thin film composite polyamide Polyether sulfone support on polyamide active layer	35.4
23	(Zhao et al., 2014)	45	poly(sodiumstyrene-4-sulfonate-co-n-isopropylacrylamide)	Low viscosity Better diffusivity	Complex design Low water flux	TFC FO membrane	Thin film composite-polyamide	4
24	(Kamio et al., 2019)	25	Thermo-responsive ionic liquids	Enhanced water flux Increased cross-flow velocity	Toxic High cost	-	-	45±7.8
25	(Pramanik et al., 2019)	40	NaCl (0.5 M)	Low concentration polarization	Low water flux after continuous use	Flat sheet thin-film composite membrane	-	28
26	(McCutcheon and Elimelech, 2007)	20	NaCl (1.5 M)	Commercially available Low cost	Enhanced fouling	Commercial membrane	-	23
27	(Hawari et al., 2016)	26	NaCl (0.5 M)	Better water flux Reduced desalination time	Low water rejection	TFC FO membrane	-	47
28	(Kim et al., 2012)	20	NaCl (5 M)	-	Low membrane transfer properties	-	-	-
29	(Lay et al., 2012)	22	NaCl (0.5 M)	Reduced ICP effects	Increased desalination time	TFC FO membrane	Thin film composite-polyamide active layer with polysulfone support layer	20
30	(X. Wang et al., 2016)	25	NaCl (1 M)	Commercially available Low cost	Increased fouling	CTA FO membrane	Cellulose triacetate	16
31	(Zhang et al., 2012)	23	NaCl (0.5 M)	Reduced biofouling	Low water recovery	TFC hollow fibre membrane	Thin film composite	22

				Better water flux				
32	(Li et al., 2012)	21	NaCl (2 M)	Low cost Reduced ICP effect	Low membrane transfer properties	CTA FO membrane	Cellulose triacetate	7.2
33	(Boo et al., 2012)	21	LaCl3	Better water recovery	Not commercially available	CTA FO membrane	Cellulose triacetate	5.3
34	(Wang et al., 2010)	23	NaCl (0.5 M)	Better water flux Better rejection rate	High cost Complex setup	cartridge-type FO flat sheet membranes	Polyester screen mesh	32.2
35	(Zeng et al., 2019)	23	-	Better recovery rate	High swelling propensity	poly(*N*-isopropylacrylamide-co-sodium acrylate) Hydrogel	poly(*N*-isopropylacrylamide-co-sodium acrylate) Hydrogel	10
36	(Altaee et al., 2013)	35	Brine	Simplistic design	Low water flux	-	-	9.6
37	(Phuntsho et al., 2013)	25	CaCl2	Enhanced water flux Low viscosity	Increased Fouling	TFC FO membrane		37.73
38	(Wang et al., 2010)	23	NaCl (4 M)	High cross-flow velocity Fouling resistant	Low durability	polyamide-polyethersulfone FO hollow fiber membranes	polyamide-polyethersulfone	25
39	(Yang et al., 2009)	23	MgCl2 (5 M)	Better water flux Reusability	Increased cost	Dual-layer polybenzimidazole-polyethersulfone (PBI-PES) nanofiltration hollow fiber membrane	polybenzimidazole-polyethersulfone (PBI-PES)	33.8
40	(Chou et al., 2010)	23	NaCl (2 M)	High rejection rate Better water recovery	Prone to biofouling	polymer polyethersulfone (PES) membrane	polymer polyethersulfone with an active layer mixture of m-phenylenediamine and tri-mesoyl chloride	42.6
41	(Ren and McCutcheon, 2014)	20	NaCl (1 M)	Better osmotic pressure Low reverse solute flux	Low water flux after continuous use	TFC FO membrane	-	22.9

42	(Qiu et al., 2011)	23	MgCl2 (3 M)	High water flux High cross flow velocity	Not commercially available	porous polyacrylonitrile (PAN) membrane	porous polyacrylonitrile	105.4
43	(Han et al., 2012b)	23	NaCl (2 M)	Simplistic design	Increased ICP effects Toxic material	polysulfone (PSf) membrane	Polysulfone with active layer mixture of m-phenylenediamine and trimesoyl chloride	14
44	(Lee et al., 2015)	23	MgCl2 (3 M)	Commercially available	Low water flux	-	-	11
45	(Emadzadeh et al., 2013)	25	NaCl (0.5 M)	Fouling resistant Enhanced water flux	High cost	thin-film nanocomposite (TFN) membrane	polysulfone–titanium dioxide	57
46	(Shakeri et al., 2019b)	25	NaCl (1 M)	High water permeability hydrophilicity	Complex design	thin film nanocomposite (TFN) membrane	Polyamide with polyester support layer	63
47	(Han et al., 2012a)	23	NaCl (2 M)	Reduced biofouling Better water flux	Increased desalination	TFC FO membrane	thin aromatic polyamide	35
48	(Ma et al., 2013)	20	NaCl (2 M)	High water flux Better membrane structure	Toxic material	Polysulfone-nanocomposite membrane	Polysulfone	86
49	(Ma et al., 2012)	20	NaCl (2 M)	Better water flux Multiple usabilities with various processes	Low water rejection	Zeolite-polyamide thin film nanocomposite (TFN) membrane	Zeolite-polyamide	48
50	(Wong et al., 2012)	30	NaCl	Low cost	Prone to fouling	CTA membrane	cellulose triacetate with polyester mesh	-
51	(Tan and Ng, 2008)	30	NaCl (2 M)	Low cost	Prone to fouling	CTA membrane	cellulose triacetate	-
52	(Bhinder et al., 2018)	23	NaCl (1.5 M)	Simple design	Low water flux	-	-	13
53	(Abdelrasoul et al., 2018)	35	NaCl	Low reverse solute flux	Increased ICP effects	TFC FO membrane	-	-

54	(McCutcheon and Elimelech, 2006)	40	NaCl (1.5 M)	Better water flux Biodegradable	Low water flux after continuous use	CTA membrane	cellulose triacetate	22
55	(Tang et al., 2010)	24	NaCl (0.5 M)	High water flux Low cost	Highly selective	CTA membrane	cellulose triacetate	57
56	(Gray et al., 2006)	22.5	NaCl (0.5 M)	Good osmotic pressure	Membrane fouling	CTA membrane	cellulose triacetate	-
57	(Suh and Lee, 2013)	23	NaCl (4 M)	Low scale desalination	Prone to fouling and scaling	-	-	18
58	(Suwaileh et al., 2019)	60	NaCl (2 M)	Better water flux High rejection rate	Complex design	Polyethersulfone (PES) UF membranes	Polyethersulfone with polyamide active layer	30
59	(Altaee et al., 2014a)	40	NaCl	Low power consumption Fouling resistant	Low cross-flow velocity Low water recovery	HTI)Hydration Technology Innovations) membrane	-	14.3
60	(Yangali-Quintanilla et al., 2011)	20	seawater	Low cost Simple design	Low desalination efficiency	-	-	10.5
61	(Bamaga et al., 2011)	25	seawater	Better osmotic pressure	Prone to fouling	CTA membrane	cellulose triacetate	11
62	(Park et al., 2018)	70	Adipic acid	Good diffusivity	Low rejection rate	CTA membrane	cellulose triacetate	-
63	(Ma et al., 2020)	25	NaCl (5 M)	Fouling resistant Good cross flow velocity	Low water flux after continuous use	CTA membrane	cellulose triacetate	15.5
64	(Zhang et al., 2019)	-	NaCl	Anti-biofouling Good dewatering capacity	Increase concentration polarization at high temperature	TFC FO membrane	-	20
65	(Liyanaarachchi et al., 2015)	40	brine	Low cost	Prone to fouling	CTA membrane	cellulose triacetate	10.2

66	(Ju and Kang, 2017)	40	zwitterionic homopolymer poly-sulfobetaine	Good water recovery Low reverse solute flux	Low water flux	TFC FO membrane	-	3.22
67	(Wang et al., 2014)	50	Cooling water	Low cost Simple design	Low water flux Low rejection rate	CTA membrane	cellulose triacetate with polyester mesh	3.1
68	(Kim et al., 2016)	25	Tetrabutyl phosphonium styrenesulfonate	Fouling resistant Better osmotic pressure	Selective effluents desalination	TFC FO membrane	Hydration Technology Innovations	16.28
69	(Yong et al., 2012)	20	NaCl (4 M)	Better water flux Lowered reverse solute flux	Increased ICP and ECP effects on prolonged use	CTA membrane	cellulose triacetate	21

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8. Summary and challenges

This section summarizes the research advances in FO technology concerning draw solution, feed solution, membranes, operating conditions and hybrid systems. The challenges and measures to address them are explained under each corresponding sub-section.

8.1. *Draw solution*

Researchers prefer DSs with low reverse solute flux and less energy consumption. Multivalent DS has low reverse solute flux and less energy consumption as compared to monovalent DS. Regenerating the DS to get freshwater is one of the main challenges and hence organic DSs are preferred, as they can be regenerated easily by volatilization.

8.2. Feed solution

In general, researchers used tapwater, groundwater and simulated industrial wastewater as the feed solution. The concentration of FS is certainly important to maintain an optimum level of concentration difference between feed and draw solution to generate the highest possible osmotic pressure gradient for maximum efficiency. Use of simulated wastewater at lab scale has been a limitation of much past research, as this may differ from actual wastewater from industries such as textile, cement and tannery industries. Hence, more recent research is tending to make greater use of real industrial wastewater from textile, manufacturing and tannery industries for experimentation.

8.3. *Membranes*

TFC membranes are preferred to treat highly concentrated wastewater from textile, manufacturing and pharmaceutical industries; whereas CTA membranes are preferred to treat less concentrated water from coal processing industries. In general, the active layer of a CTA membrane is modified to improve the surface morphology, selectivity, hydrophilicity, and structural stability; whereas the selective layer is modified to enhance the membrane properties like stability, tortuosity, mechanical strength, porosity and

permeability, the support layer should be modified. Nanoparticles can enhance all these properties further. Fouling is one of the main challenges, which affects the membrane efficiency. Even though fouling can be minimized by membrane cleaning, its initial efficiency cannot be restored after cleaning. Hence, effective cleaning techniques are desirable to restore the initial membrane efficiency.

8.4.*Operating conditions*

Water flux can be improved by maintaining a constant temperature in both DS and FS due to the enhanced mass transfer across the FO membrane. Increasing the flow rates of DS and FS simultaneously reduces the concentration polarization across the FO membrane and improves the water flux. Modifying the flow rate of DS and FS reduces the ECP, whereas support layer modification reduces the ICP in the FO membrane. AL-FS membrane orientation is preferred to treat hypersaline water, whereas AL-DS was preferred to treat low saline water. Maintaining the constant temperature of feed or draw solution is one of the main challenges for applications in temperate climate zones, where the ambient temperature will be less than 15℃. Heating may be needed to maintain the temperature of FS and DS, leading to additional energy consumption. Hence, there is on-going research for overall process optimisation with respect to effectiveness and energy consumption.

8.5.*Hybrid systems*

FO systems may be integrated with other technologies like RO and MED to reduce the specific energy consumption and to improve the overall salt rejection rate, water permeability and flux. High energy consumption is one of the main challenges in integrating FO with other RO and MED. Hence, energy consumption could be further reduced by the integration of FO with waste heat recovery devices.

9. Conclusions and future research prospects

Over the past two decades (from 2000 to 2021) there has been a dramatic increase in research and development in forward osmosis. Based on the ScienceDirect database, there was around a seven-fold increase in the total number of publications on FO between 2000 to 2010, followed by a further 350% increase from 2010 to 2019. The main areas of research include membrane fabrication and modification, novel DS and regeneration methods to reduce specific energy consumption, fouling reduction techniques and optimization of operating and process parameters.

According to Gluck (Gluck, 2017), FO is still at an early commercial stage. Though the number of installations worldwide is small, however, the number of companies active in this area is increasing. BlueTech was among the first companies active in this field, with activities reported up to 2015. Since then other companies like $FTSH_2O$, Porifera, Oasys and Modern Water and Aquaporin have become prominent. The number of installations, however, still only totals about 50 worldwide. Despite its early stage of development, in coming years FO will likely become commercialised in various industries including textile, dairy and tannery to treat wastewater effectively.

In this review, we have discussed membrane developments, novel DS, changes in system temperatures, and hybrid systems targeted at increasing the efficiency of FO. Nevertheless, many DS's including hydro-acid complexes, super-hydrophilic nanoparticles and recently-developed nanoparticles, such as graphene and carbon nanofibers, are still in their infancy as regards use in FO. Hence, research and development in these areas are expected to provide further enhancements to water flux.

Research should also be directed towards more efficient ways of integrating PRO with FO to enhance the efficiency and water flux. Concerning membrane materials, novel membranes like mesoporous silica material-15, 16, and graphene oxide – which possess high hydrophilicity with low solute diffusion, fouling, ICP and ECP – can be used in FO.

Concerning the draw solution, effective regeneration methods which consume less energy must be identified. Future research can be focussed on integrating shape memory polymer (SMP) with hydrogel as an effective DS as this may have various advantages like restoring the original shape under heat or light. Optimization studies employing a multi-criteria decision-making process should be carried out to optimize the concentration of FS, DS and membrane to arrive at a maximum water flux. It is important that future lab studies should use real industrial wastewater rather than only simulated wastewater.

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