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Studies on performances of membrane, draw solute and modeling of forward osmosis process in desalination – a review

Sobana Subramani, Rames C. Panda*, Bidisha Panda

Chemical Engineering Department, CSIR-CLRI, Adyar, Chennai 600 020, India, Fax: +91 (44) 24911589; emails: panda@clri.res.in (R.C. Panda), sobbana.subramani@gmail.com (S. Subramani)

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ABSTRACT

Forward osmosis (FO) is a membrane process that makes use of the osmosis phenomenon for the transport of water from a feed solution to a draw solution across a highly selective membrane under a driving force provided by the osmotic pressure difference between the two solutions. Based on energy consumption, this technology has got an edge over others. However, limited advancement on theoretical modeling for prediction of performance, lack of an ideal FO draw solution, concentration polarization and lack of economic feasibility have made this technology to proceed through further research with an objective of commercialization from 1970 to 2016. Although the technology has many potential applications, like wastewater treatment, membrane bioreactor, oil and gas, pharmaceutical, food and beverage etc., it still faces considerable limitations, including concentration polarization, membrane development and characterization, reverse solute diffusion, development of draw solution and their recovery. In order to address these issues more research is required. This paper presents a state-of-art review on FO technology covering types of membrane, draw solute, their characteristics, concentration polarization, identification of parameters, dynamic modeling of separation, novel membrane and hybrid systems from 103 literatures.

Keywords: Modeling; Membrane; Concentration polarization; Identification; Forward osmosis

1. Introduction

Water scarcity due to global warming (less rainfall) and seasonal anomaly has led process industries to reuse utility (water) or to extract water from sea to a maximum extent. Freshwater shortage in future will be unavoidable, and most of the world water reserve is in the form of seawater. Reuse of process effluents will not only reduce the demand for freshwater but also minimize pollution and emission loads. The effluents of wastewater from all process industries have been reduced through desalination technologies [1]. There is an urgent need for developing more efficient desalination techniques for solving the long-term need of water from desalination. This is also a long-term objective for researchers in the field of desalination. Generally desalination is done by distillation (multi-stage flash [MSF], multiple effect, evaporation, vapor compression) or membrane processes (electrodialysis reversal, nano-filtration [NF], membrane distillation [MD], forward osmosis [FO] and reverse osmosis [RO]). Out of all these, RO using cellulose acetate (CA) or polyamide membranes is widely used [2]. However, disposal of rejected salt and the cost of production due to requirement of high pressure (non-energy efficient techniques) limit it from practical use. Moreover, the RO process uses hydraulic pressure as the driving force for separation, which serves to counteract the osmotic pressure gradient that would otherwise favor water flux from the permeate to the feed. RO needs higher applied pressure (the driving force for mass transport). Out of the reports on the likely low-energy desalination processes, FO has been reported to consume the least energy for desalinating seawater [3]. In addition to waste heat, the electrical energy needed is about 0.25 kWh/m^3. Based on the energy consumption, the FO desalination process

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* Corresponding author.

consumes only one-third of energy required compared with the RO and one-fourth of energy compared with the MSF thermal desalination processes.

Hence, FO is gaining more attention, as it is an energy-efficient technique [4]. However, due to scaling and fouling, membranes degrade that decreases the effectiveness of the membranes in reducing the solids demanding more frequent cleanings. That means deposition of unwanted cakes on the membranes increases resistance of membrane requiring higher operating pressure but lower permeate rate. Elimelech [5] gave details of a novel FO desalination process that is being investigated and developed at Yale University in New Haven, Connecticut, and suggested that the FO technology promises lower energy costs, higher recovery and less brine discharge than conventional desalination systems [5,6]. A scale on a membrane provides nucleation sites that increase the rate of formation of additional scale [7]. Coagulation, clarification and associated unit process during pre-treatment/primary treatment helps (by neutralizing the negative surface of the suspended solids) in conglomeration and precipitation of suspended/dissolved solids that in turn increase the problems of RO membranes. FO is becoming popular as it helps to address all these issues.

It (FO) is a membrane process that has been studied in recent years as novel technology for treatment of a wide variety of aqueous solutions. FO is an osmotic process that, similar to RO, uses a semi-permeable membrane to effect separation of water from dissolved solutes. The driving force for this separation is an osmotic pressure gradient, such that a solution (A) of high concentration (relative to that of the feed solution (B) is used to induce a net flow of water through the membrane into the solution, thus effectively separating the feed water (FW) from its solutes. In the FO seawater desalination process, the feed solution is the seawater having lower osmotic pressure. The high salinity solution performs as the draw solution, which has a higher osmotic pressure than the feed solution, to induce water flow across the membrane from the feed solution to itself. The draw solution provides high osmotic pressure due to high concentration of solute. The solute from the seawater diffuses through the membrane toward the draw solution and causes dilution to the draw solution. Clean water is extracted from the seawater, and the diluted concentrated draw solution is recycled [8]. This process has immense potential for seawater desalination due to its promising low energy consumption. The energy consumption for FO process is only 2 kWh/m^3 considering 50% recovery compared with RO, where the energy consumption is 5 kWh/m^3. The main advantages of using FO are that it operates at low or no hydraulic pressures; it has ability of high rejection of a wide range of contaminants; and it may have a lower membrane fouling propensity than pressure-driven membrane processes [9].

There are many reports on development of membrane for FO that are fabricated by traditional phase inversion and thin-film composites (TFCs) via interfacial polymerization methods [10]. The reverse salt fluxes of these membranes are high that can be made usable by introducing hydrophilic materials as substrates. The mechanical properties of the membranes can be further improved by introducing nano-fiber and multi-bore substrates. Recently, double-skinned FO membranes, consisting of a dense RO layer and a loose RO layer, have been used to reduce fouling and internal-concentration-polarization problems [11]. Most of the works reported on the development of FO desalination system are related to laboratory experiments and process simulations. Moreover, technology relating to internal/external concentration polarization (ICP/ECP) that lowers the flux of permeate, the choice of draw fluid and its regeneration and development of suitable commercial FO membranes is necessary at present. Loeb [12] proposed large-scale plants for handling seawater. Recently, Bruggen and Luis [13] reviewed FO applied for production of potable water and wastewater treatment. Achilli et al. [14] discussed reverse draw solute (DS) flux, concentration polarization and the regeneration of the draw solution in FO. Qin et al. [15] presented recent developments and future challenges of FO for desalination by reviewing 76 articles where they discussed roles of dilutive ICP and concentrative ECP on the limitation of FO membranes. Akther et al. [16] gave a comprehensive review on FO highlighting improvements in FO membrane permeability and regenerative DSs including major challenges, limitations and hindrances of FO activity since 2009. Chekli et al. [17] and Eyvaz et al. [18] also reviewed developments on membrane bioreactors using FO for treating wastewater. They also explored possibilities of using hybrid system with FO followed by RO. After designing FO plant, the next step is to think for safe operation and control through monitoring process variables regularly using proper instrumentation. Reimund et al. [19] reviewed FO and discussed energy efficiency, membrane properties and benefits/applications in different processes. The review did not include identification and control aspects in the operation of FO systems [20,21].

From the state of art discussed above, it is felt that there is lack in understanding the mechanism behind the process. Not many comprehensive transient models [22] incorporating internal/ECP of solute components, hydraulics and dynamics of brine rejections etc. exist today that are needed for proper operation and control of the process. Transport mechanism inside the membrane, membrane fouling and maintenance need to be found.

To design a typical FO effectively and efficiently, thorough studies on this process using/through models are necessary. This study will help in finding safe operation strategies and in achieving better performance of the process. Thus, the entire paper is organized as follows: section 2 describes principle of operation of FO. Characterization of membranes and DS are discussed in section 3. Modeling of FO and development of concentration polarizations works are presented in section 4. Section 5 summarizes available identification techniques of process parameters of FO in order to construct an easy model. Application of FO technique and hybrid processes are discussed in section 6. FO coupled with a MD is discussed also in this section. Section 7 discussed the comparison of RO and FO. Control aspects for safe operation of the technique are explained in section 8. Future direction of research has been discussed in section 9. At the end conclusion has been drawn from the study.

2. Principle of operation

Osmosis is the flow of solvent, generally water, across membrane. The driving force for the flow is the difference in chemical potential on the two sides of the membrane. The solvent is allowed to flow from the region of higher potential to the lower potential. In practice, osmotic pressure of solution is the pressure that must be applied to the solution to arrest net flow from pure solvent across the membrane to the solution. Using Van 't Hoff equation osmotic pressure is directly proportional to the concentration of solutes.

$$\Pi = nRT \tag{1}$$

where n is the sum of all ions in solution; R is the ideal gas constant; and T is the absolute temperature in Kelvin.

The effect of temperature on a saturation concentration of binary salt can be calculated by using the relation developed by Miller and Evans [23]:

$$\begin{aligned} \Delta G &= \Delta H - T\Delta S \\ \Delta G &= RT\ln K \\ K &= \left[A\right]\left[B\right] \end{aligned} \tag{2}$$

where G, H and S are Gibbs free energy entropy and enthalpy, respectively; K is the solubility product; and A and B are the molar concentration of ion A and B in the solution. Given the thermodynamic parameters of dissolution process, ion concentration in a saturated solution of AB and the resulting osmotic pressure as a function of temperature can be calculated. The osmotic pressure of sea as a function of temperature can be calculated as at 40°C with 9.6 π atm of saturated salt concentration; the osmotic pressure becomes about 31 atm [24]. The draw solution of high concentration (relative to that of the feed solution) induces a net flow of water through the membrane into the draw solution, thus effectively separating the FW from its solutes. In contrast, the RO process uses hydraulic pressure as the driving force for separation, which serves to counteract the osmotic pressure gradient that would favor water flux from the permeate to the feed. Fig. 1 shows the schematic representation of an FO osmotic setup.

FO is a promising tool that uses CA or polyamide membrane [25] and is widely chosen separation technology as the cost of production is reduced by the use of energy efficient techniques.

3. Membranes and draw solutions

Fig. 2 shows the schematic diagram of the lab-scale FO setup with co-current flow of feed NaCl where draw solution of ammonium–carbon dioxide is used in the cross-flow cell. Computer monitors the mass of water permeating into the draw solution, from which product water flux is calculated.

The active layer of the membrane is generally made of cellulose triacetate. The support is made of a woven fabric mesh embedded with the porous support portion of the membrane and is of asymmetric structure as given in references [24,26]. Low [27] suggested that this structure may increase the thickness of the FO membrane and hinder the flow of permeate. The performance of this asymmetric membrane is poor due to boundary layer that occurs within the supporting layer for osmotic driven pressure as pointed out in references [26,28,29] where the latter discussed that the asymmetric membrane structure creates an ICP effect. Depending upon the membrane orientation, two types of ICP exist as given by Moody and Kessler [30].

In one orientation, the feed solution is directed against the support layer which is known as pressure-retarded osmosis (PRO) mode. In other orientation where the draw solution is directed against the support layer, is known as FO mode. In both the modes, ICP occurs in the porous support layer. Concentration polarization on the feed side of the membrane inhibits permeate flux as given in references [31,32]. This phenomenon occurs due to the increased osmotic pressure at the membrane active layer interface on the feed side of the membrane. This is known as ECP. This ECP on the feed and permeate side is known as concentrative and dilutive ECP, respectively. Lab-scale FO membrane setup is shown in schematic diagram (Fig. 2 and Tan and Ng [33]). The flux of sucrose is large compared with ammonium carbonate, though the sucrose has heavier molecular weight and lower osmotic pressure difference. Compared with ammonium–carbon dioxide flux, ammonium carbonate produces fluxes at 40% higher rate.

Benko et al. [28] reported an apparatus to measure unsteady flux of pure water through combined permeable selective RO and NF membranes under an osmotic pressure gradient (aka FO), which affect the performance of water permeation and solubility.

Wang et al. [34] proposed polybenzimidazole (PBI) NF membrane, having narrow pore size distribution membrane, for FO using $MgCl_2$ solutions at different concentration and other salt solutions separately as draw solutions to achieve high water permeation flux and excellent salt selectivity.

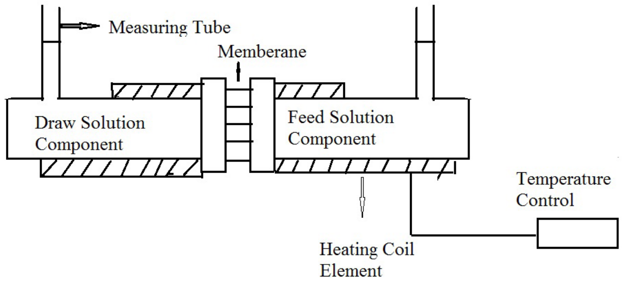

Fig. 1. Schematic representation of an FO osmotic setup.

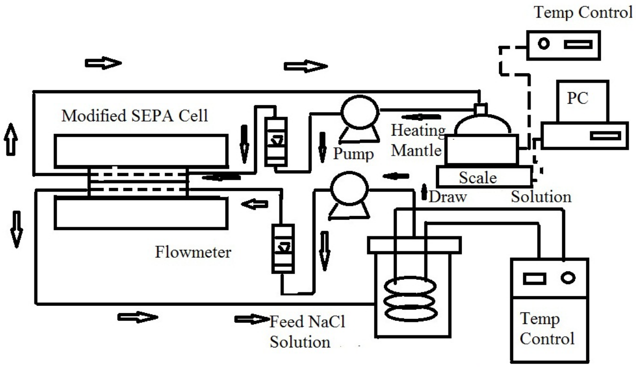

Fig. 2. Schematic diagram of the lab-scale forward osmosis setup with co-current flow of feed NaCl and draw solution of ammonium and carbon dioxide.

McCutcheon et al. [35] studied FO and PRO and proposed that water flux depends on the utilization of large osmotic pressure differentials across semi-permeable membranes made up of polyester, poly ethylene terephthalate (PET) non-woven and polysulfone support layer. They also showed effects of structural characteristics of asymmetric membrane on the performance of water flux. Lu et al. [36] used the ability of NaCl and dendrimers as draw solutions to produce highest osmotic pressures, to enhance membranes characteristics in FO. They highlighted FO technique as minimum cost-effective option compared with RO concentration disposal. Yang et al. [25] fabricated dual-layer PBI–polyethersulfone (PBI–PES) NF hollow fiber membranes via co-extrusion technology consisting of ultrathin selective porous channels supported by a microporous sponge-like structure to carry water in the FO process for producing potable water. In a separate attempt, Wang et al. [34] fabricated PBI NF hollow fiber FO membranes with high flux. The separation of salt from seawater was achieved via non-solvent phase inversion and modification through chemical cross-linking. The cross-linking characteristic of *p*-Xylylene dichloride was used to tune the mean pore size to enhance the salt selectivity for improving flux of water. The 2-h modified PBI NF membrane can be reused for water salt selectivity [37].

Yip et al. [26] prepared another technique selective polyamide active layer by interfacial polymerization on top of a polysulfone support layer fabricated by phase separation onto a thin (40 μm) polyester non-woven fabric as FO membrane. Polymer concentration and solvent composition was mixed to make polysulfone casting solution to support a layer containing mix of finger-like and sponge-like morphologies for FO using 1.5 M NaCl as draw solution and pure water as feed. The produced membranes gave permeate flow of water exceeding 18 L m^{-2} h^{-1}, along with salt rejection greater than 97%. The output depends on porosity, thickness, tortuosity and pore structure of the polysulfone support layer. On application, the membrane showed good performance with ammonium bicarbonate as draw solution [38]. Su et al. [39] annealed the microstructure and decreased the mean pore radius from 0.63 nm to 0.39, 0.36, 0.30 and 0.30 nm at 70°C, 80°C, 85°C and 90°C, respectively, which were used to study the effects of CA hollow fiber FO membrane. As a result of annealing, the sizes of free volume of the FO membrane decreased in the depth range of 0.31–1.33 μm at 80°C. Yen et al. [40] studied effect of 2-methylimidazole-based organic compounds as DSs and compared results between the neutral and charged compounds in FO membrane. The charged DSs A and B exhibited higher water flux performance and lower reverse solute fluxes than the neutral DSs C and D, which were based on hydration technology innovation (HTI) cellulose triacetate membranes, and the solutes A and B showed similar water fluxes even though the latter had a higher osmotic pressure [26]. Jia et al. [38] considered neutral armchair (*R*,*R*) on carbon nano-tubes (CNTs) with indexes $R = 6 - 11$ to study the transport behavior of water molecules and ions through the membranes by molecular dynamic simulation. The membrane incorporating CNT (8,8) achieved optimum salt rejection. Fourteen draw solutions were selected to evaluate the FO membrane performance in water flux and reverse salt diffusion [39,40]. CA NF hollow fiber FO membranes were fabricated using two-step heat treatment (i.e., for 60 min at 60°C and for 20 min at 95°C), which shrunk the membrane mean pore radius from 0.63 to 0.30 nm [41]. Average molecular weight of the resultant CA–NF membrane was 186 Da under 1 bar transmembrane pressure that showed pure water permeability of 0.47 L/m^2 bar h and rejection levels of 90.17% for NaCl and 96.67% for $MgCl_2$. Zhang et al. [41] explored the FO membrane design and engineering of membrane structure that gave low salt leakage and minimized ICP. Tiraferri et al. [42] investigated the performance of the influence of the TFC membrane support layer structures consisted of a selective polyamide active layer formed by interfacial polyamine on top of a polysulfone support layer fabricated by phase separation. Separation of microalgae was achieved from algal biodiesel production where the physical and chemical parameters affecting FO flux performance were investigated systematically [43]. They found that the effect of solute reverse diffusion on FO fouling was more severe on draw solution concentrations and on the active-layer-facing-the-draw-solution orientation. Magnetic nano-particles (MNPs) were synthesized, using coating with a series of poly(ethylene glycol) diacid-coated (PEG-$(COOH)_2$-coated) particles of different size distributions [44]. The physical and chemical properties of the resultant MNPs are analyzed by TEM and are fully exploited for their recovery and reusability in FO systems. In order to overcome the ICP effect that induced hindrance on (1) separation (salt flux) and (2) the performance (water flux), new FO membranes were fabricated using a highly porous sublayer sandwiched between two selective skin layers via phase inversion [45]. Ethylenediaminetetraacetic acid disodium (EDTA-2Na) salt was used as novel draw solution for investigating dewatering phenomena of high nutrient sludge using FO membranes made of flat-sheet cellulose triacetate and with effective membrane area of 92.95 cm^2 [46]. FO membrane was able to separate the higher permeate flux and lower reverse solute flux as compared with conventional inorganic salt (i.e., NaCl, $MgSO_4$). In a separate attempt, the nutrients and organic compounds in activated sludge were also successfully removed. Considering salinity as potential gradient between seawater and reclaimed water, Cath et al. [47] designed forward osmotic dilution of seawater. Four major advantages in their approach are: lower energy desalination of seawater, multi-barrier protection of drinking water, reduction in RO membrane fouling due to impurities in impaired water and beneficial reuse of impaired water. Superhydrophilic, citrate-coated MNPs (cit-MNPs) were synthesized through a co-precipitation method and were analyzed for their special characteristics (highly negative surface charge and high surface charge density) and a rapid decline in the water flux occurred due to an interaction between the cellulose triacetate membrane and cit-MNPs [48]. Hydrogels made of co-polymer microgels of *N*-isopropylacrylamide and acrylic acid with different mixing ratios were synthesized using surfactant-free emulsion polymerization to produce submicron-size hydrogels with high surface area, and fast swelling/deswelling response was employed as draw agents for FO process [49]. Tian et al. [50] developed draw solution made of poly(sodium 4-styrenesulfonate) (PSS) polyelectrolytes with different molecular weights with different concentrations, which showed that conductivity of permeate and the osmotic pressure increase with the increase of PSS concentration. It was also found that the polyelectrolyte with higher molecular weights generated severe concentration polarization in FO process. A novel DS, namely EDTA-2Na coupled with Triton X-100 was proposed by Nguyen et al. [51] to achieve zero diffusion and deionized (DI) water as feed solution in FO application. Long and Wang [52] proposed a DS sodium tetraethylenepentamine heptaacetate, which was characterized by nuclear magnetic resonance spectroscopy and Fourier transform infrared. The optimum pH and concentration of feeds were used for calculation of performance of FO. *Pseudomonas aeruginosa* was embedded in pristine and chlorine-treated TFC polyamide membranes with either $MgCl_2$ or $CaCl_2$ as draw solution to prevent biofouling [53]. FO membranes were grafted with poly(ethylene glycol) (PEG) block co-polymer onto the surface of commercial TFC to increase membrane hydrophilicity with reduced water permeability [54]. The performance of this method was compared with TFC–FO membranes, and it was claimed that this new method is relatively inexpensive, easy to implement and applicable to commercial membranes. Table 1 presents a comparative study on the state of art on use of DS to increase the flux with decreased solubility. The main challenge regarding the selection of appropriate DS is their separation and recovery from the product water, especially for application to drinking water. Thermodynamic software providing the properties of solute (library containing data on including water solubility, pH, speciation and osmotic pressure) is helpful in selection of DS. A DS with inert, harmless or less toxic and with near neutral pH may be suitable for preparation of good drinking water. FO membrane should also be economic, compatible with DS and should not impart unwanted fouling, reaction, adsorption or dissolution. Many researchers [55–57] have presented studies on effects of parameters (DS characteristics and operating conditions) on performance of FO. Solute resistance (K_S) to diffusion can be related to diffusion constant (D_S) as $K_S = \frac{t_p \tau_p}{\phi D_s}$ where t_p is thickness; τ_p is tortuosity; and ϕ is porosity of solute [55]. With high DS concentration, water flux changes with logarithmic [56]. Effect of ICP hinders higher permeate flux resulting in less effective water flux [57]. Achilli et al. [14] used improved DS containing scale precursor ions for food industry. Novel DS containing MNPs have been developed by Ling et al. [58] that helps to aggregate unwanted particles and enhance membrane activity.

Above developments of characteristics of the DS motivates to concentrate on formulation of models and identification of their respective parameters in the scale-up of FO systems.

4. Mathematical modeling

Models that adequately describe the performance of FO membranes are very important since these are needed in the design of FO processes. Moody and Kessler [30] discussed the relationship between the kinetics and design parameters of FO to apply FO process to problems such as agricultural water reclamation, dehydration of solutions and the production of potable nutrient solutions from seawater. Three mathematical models were developed that include increasingly complex assumptions, are presented and in all cases the fundamental mechanical device is assumed to be a continuous flow extractor that incorporates a semi-permeable membrane. The solution diffusion model predicted that with decreasing osmotic driving force in FO process, the water flux decreases proportionally irrespective of the feed or draw solution concentration [62]. The direction of water flux is opposite to the diffusion of DS [63]. However, they suggested that feed solution concentration may be an important factor that influences the permeate flux in FO process. With the experimental data they showed that for similar osmotic pressure difference between bulk feed and draw solution, higher concentration feed produces less flux as suggested by Loeb et al. [64] that orientation of membrane has influence on water flux. Models that predict separation characteristics also minimize the number of experiments that must be performed to describe a particular system.

McGinnis et al. [65,66] proposed closed cycle PRO process (i.e., osmotic heat engine) to generate osmotic power from seawater using a concentrated ammonia–carbon dioxide as draw solution and suggested that most difficult problem in modeling FO is the ICP, which reduces water flux by up to 80%. In a PRO process, power density (W) is normalized by the membrane area (e.g., m^2) and may be given by:

$$W = J_W \Delta P \tag{3}$$

where ΔP is the hydraulic pressure different across the membrane, and Jw is the water flux, which can be expressed by Eq. (2):

$$J_W = A(\Delta\pi - \Delta P) \tag{4}$$

where A is the water permeability coefficient of the membrane, and $\Delta\pi$ is the osmotic pressure differential across the membrane. Combining Eqs. (3) and (4), they showed:

$$W = A(\Delta\pi - \Delta P)\,\Delta P = -A(\Delta P - \Delta\pi/2)^2 + A\Delta\pi^2/4 \tag{5}$$

McCutcheon et al. [35] presented the modified flux equation for higher flux rate by including both the concentrative and dilutive ECP as:

$$J_W = A\left[\pi_{d,b}\exp\left(\frac{-J_W}{K}\right) - \pi_{f,b}\exp\left(\frac{J_W}{K}\right)\right] \tag{6}$$

Dense symmetric membrane is not in use today; therefore, the use of the above model is restricted. If the membrane is asymmetric, then by coupling concentrative ICP with draw ECP, an analytical model was derived to predict the permeate flux by placing the feed against the support layer and the draw solution against the active layer given by McCutcheon et al. [35]:

$$J_W = A\left[\pi_{d,b}\exp\left(\frac{-J_W}{K}\right) - \pi_{f,b}\exp\left(J_W K\right)\right] \tag{7}$$

The draw solution was placed against the support layer and the feed against the active layer for an asymmetric membrane, and another analytical model was derived to predict the flux as:

Table 1
Comparison of different draw solutes with their flux and solubility as studied by different authors

S. No.	Author	Year	Name of the feed solution	Name of the draw solution	Increase of flux	Decrease of solubility
1.	McCutcheon et al. [59]	2005	0.5 mol of NaCl	6 mol of ammonium bicarbonate salt (NH_4HCO_3) in water	10 μm/s at 50°C	95%–99%
2.	Wang et al. [34]	2007	DI water	$MgCl_2$	20 kg/m^2 h	92.7%
3.	Mi and Elimelech [60]	2008	50 mM NaCl	0.4 M NaCl and dendrimers	8 μm/s	93%
4.	Low [27]	2009	NaCl	Sucrose		
5.	Yip et al. [26]	2010	Water	1.5 M NaCl	18 L/m^2 h	97%
6.	Yen et al. [40]	2010	Water	2-Methylimidazole-based organic compounds, 1–4 as draw solution	20 L/m^2 h	0.4/5 M
7.	Ling et al. [44]	2010	Water	Poly(ethylene glycol) diacid-coated (PEG-$(COOH)_2$-coated) MNPs	5 L/m^2 h	0.8 mol/L
8.	Jia et al. [38]	2010	Water	Neutral armchair (*R*,*R*) C nanotubes (CNTs) with indexes *R* = 6–11, CNT (11-11)	Water molecule number 40/s	3.04/5 mol/L
9.	Ling et al. [58]	2011	Water	Magnetic nanoparticle solution		
10.	Zou et al. [43]	2011	Bioalgae	Ammonia	Separation of microalgae	
11.	Hau et al. [46]	2014	High nutrient sludge	Dewatering of ethylenediaminetetraacetic acid disodium (EDTA-2Na) salt	Cross-flow rate of 6.4 cm/s	Final sludge concentration obtained was 34,700 mg/L
12.	Na et al. [48]	2014	Pure water	Superhydrophilic, citrate-coated magnetic nanoparticles (cit-MNPs)	17.3 L/m^2 h with 20 mg/L MNP	
13.	Zaviska and Zou [31]	2014	Real brackish groundwater	$Na_2SO_{4,}$ NaCl, $MgSO_4$	22 L/m^2 h, 17.8 L/m^2 h, 14.5 L/m^2 h,	Not reported
14.	Sato et al. [61]	2014	NaCl aqueous solution	Ether	2.12–2.91 L/m^2 h	Not reported
15.	Hartanto et al. [49]	2015	Water	*o*-Polymer microgels of *N*-isopropyl acrylamide and acrylic acid with different mixing ratios were synthesized using surfactant-free emulsion polymerization to produce submicron-size hydrogels	23.8 L/m^2 h (72.4%)	
16.	Tian et al. [50]	2015	0.5 mol/ L NaCl	Poly(sodium 4-styrenesulfonate) (PSS) polyelectrolytes with different molecular weights and different concentrations	Higher molecular weight will yield lower flux	
17.	Nguyen et al. [51]	2015	Deionized water	0.1 M EDTA-2Na coupled with 0.05 mM Triton X-100	4.6 L/m^2 h	95%
18.	Long and Wang [52]	2015	Deionized water	0.5 g/mL sodium tetraethylenepentamine heptaacetate	28.57 L/m^2 h	0.45 g/h

$$J_W = A\left[\pi_{d,b}\exp\left(-J_W K\right) - \pi_{f,b}\exp\left(\frac{J_W}{K}\right)\right] \tag{8}$$

In each of the above cases, ECP and ICP module contributed negatively to the overall osmotic driving pressure. The above two models were verified experimentally. The modeling of these relationships is in practice more complex than this equation indicates, with flux depending on the membrane, feed and draw solution characteristics, as well as the fluid dynamics within the process itself as pointed out by Lee et al. [67]. A steady-state mass balance can be written on a differential volume for the support layer given by McCutcheon and Elimelech [22] as:

$$\frac{dJ_s^S}{dz} = -D^S \frac{d^2C}{dZ^2} + J_W \frac{dC}{dZ} = 0 \tag{9}$$

where J_s^S is the total flux of DS; C is the solute concentration; $D^s = \frac{D\varepsilon}{\tau}$, which is the solute diffusion coefficient in the support layer; and J_w is the superficial fluid velocity (solvent permeate flux).

The DS concentration profile in the support layer is also given by the expression as in [68]:

$$C = \frac{\exp\left(\frac{J_W t_S \tau \frac{Z}{t_S}}{D\varepsilon}\right)\left(C_D - C_i^S\right) + \exp\left(\frac{J_W t_S \tau}{D\varepsilon}\right) C_i^S - C_D}{\exp\left(\frac{J_W t_S \tau}{D\varepsilon}\right) - 1} \tag{10}$$

where C_i^S is the DS concentration on the support layer side of the support layer–active layer interference; C_D is the bulk DS concentration; and t_S is the support layer thickness. The solute flux into the feed solution is derived as follows:

$$C = \frac{\exp\left(\frac{J_W t_S \tau}{D\varepsilon}\right) C_i^S - C_D}{\exp\left(\frac{J_W t_S \tau}{D\varepsilon}\right) - 1} \tag{11}$$

Analytical expression for the reverse flux for the DS is given as follows:

$$J_S = \frac{J_W C_D}{1 - \left(1 + \frac{J_W t_A}{D^A H}\right)\exp\left(\frac{J_W t_S \tau}{D\varepsilon}\right)} = \frac{J_W C_D}{1 - \left(1 + \frac{J_W}{B}\right)\exp\left(\frac{J_W S}{D}\right)} \tag{12}$$

where H is the partition coefficient describing the relative concentration in each phase. Membrane's structural parameter $(S) = \frac{t_S \tau}{\varepsilon}$ characterizes the average distance traveled by a solute molecule through a support layer, while going from the bulk draw solution to the support side. Active salt permeability coefficient $(B) = \frac{D^A H}{t_A}$. J_s and the ratio of $\frac{J_w}{J_s}$ were determined experimentally. Moody and Kessler [30] proposed the asymmetric membrane flux modeling where the value of solute resistance to diffusion (K) was varied to determine the impact of membrane design on flux performance. A better flux performance was predicted by reducing the value of K. Though the flux improvement was limited by ECP, the better flux performance is obtained due to reduced severity of ICP on membrane orientation as suggested by Moody and Kessler [30].

McCutcheon and Elimelech [22] and Gray et al. [69] proposed film theory where the concentration of solute at the membrane interface is given as:

$$J_W = -D\frac{dC}{dX} \tag{13}$$

Integrating Eq. (6):

$$J_W = -K \ln \frac{C_{d.w}}{C_{d.b}} \tag{14}$$

where K depends strongly on the hydrodynamics of FO process, which can be related to the Sherwood number as:

$$K = \frac{ShD}{d_h} \tag{15}$$

Mulder [63] modeled ECP effect for RO, ultra filtration (UF) and FO process using equations consisting of Sherwood number as:

$$Sh = 1.85\left(\mathrm{Re}\, Sc \frac{dh}{L}\right)^{0.33} \text{ for laminar flow } (\mathrm{Re} \le 2100) \tag{16}$$

$$Sh = 0.04\,\mathrm{Re}^{0.75} Sc^{0.33} \text{ for turbulent flow } \left(\mathrm{Re} > 2100\right) \tag{17}$$

where Re is the Reynolds number.

But the Sherwood relation for UF and FO are different because of the finer membrane surface and the limited channel length and negligible pressure drop along the channel length of FO membrane. By taking the above points into consideration and by using boundary layer concept, an exact solution was developed for local mass transfer coefficient for hydrodynamic boundary layer of fluid that flows parallels to a smooth, non-porous and flat surface as given by Welty et al. [70], which correlated the boundary layer concept with the primary local Sherwood number as:

$$\text{Laminar boundary layer } \left(\mathrm{Re}_y \le 2\times10^5\right)\ \mathrm{Sh} = 0.332\ \mathrm{Re}_y^{\,1/2}\ \mathrm{SC}^{1/3} \tag{18}$$

$$\text{Turbulent boundary layer } \left(\mathrm{Re} > 2\times10^5\right)\ \mathrm{Sh} = 0.029\mathrm{Re}_y^{\,4/5}\mathrm{SC}^{1/3} \tag{19}$$

The mean mass transfer coefficient (K_c) was obtained as:

$$K_c = \frac{\int_0^L K_{dy}}{\int_0^L dy} \tag{20}$$

$$K_C = \frac{0.664D(\mathrm{Re}_t)^{\frac{1}{2}}\left(SC\right)^{\frac{1}{3}} + 0.0365D\left(SC\right)^{\frac{1}{3}}\left[\left(\mathrm{Re}_L\right)^{\frac{4}{5}} - \left(\mathrm{Re}_t\right)^{\frac{4}{5}}\right]}{L} \tag{21}$$

where K is the resistance to solute diffusion within the membrane porous support layer. Loeb [12] and Loeb et al. [64] derived the equation for K as:

$$K = \left(\frac{1}{J_W}\right)\ln\left[\frac{B + A\Delta\pi}{B + J_W + A\Delta P}\right] \text{for dilutive ICP and} \tag{22}$$

$$K = \left(\frac{1}{J_W}\right)\ln\left[\frac{B + A\Delta\pi - J_W}{B + A\Delta P}\right] \text{for concentrate ICP.} \tag{23}$$

where A and B are the water and solute permeability coefficient of the active layer of the membrane, respectively. K values for various solutes are found to be inversely proportional to the diffusion coefficient of the solution as given by Na et al. [48]:

$$K = \frac{t\tau}{\varepsilon D_S} \tag{24}$$

where t is the membrane thickness; τ is the tortuosity of the membrane porous support layer; ε is the porosity of the porous support layer; and D_s is the diffusion coefficient of solute.

An expression for modeling the dilutive ICP and concentrated ICP was developed using the governing equation, which is similarly derived by Loeb et al. [64] as follows. Tan and Ng [33] proposed formula for resistance to solute diffusion as:

$$K^* = \left[\frac{F_1}{J_W}\left(C_{d,b} - C_{d,w}\right) + \frac{F_2}{J_W}\left(C_{d,b}^2 - C_{d,w}^2\right) + \ldots.. \frac{F_n}{J_W}\left(C_{d,b}^n - C_{d,w}^n\right)\right] + \frac{G}{J_W}\ln\left[\frac{B\left(C_{d,w} - C_{f,w}\right) + J_W C_{d,b}}{B\left(C_{d,w} - C_{f,w}\right) + J_W C_{d,w}}\right] \tag{25}$$

$$K^* = \frac{t\tau}{\varepsilon} \tag{26}$$

$$K^* = \left[\frac{F_1}{J_W}\left(C_{d,w} - C_{db}\right) + \frac{F_2}{J_W}\left(C_{d,w}^2 - C_{d,b}^2\right) + \ldots.. \frac{F_n}{J_W}\left(C_{d,w}^n - C_{d,b}^n\right)\right] + \frac{G}{J_W}\ln\left[\frac{B\left(C_{f,w} - C_{d,w}\right) + J_W C_{d,w}}{B\left(C_{f,w} - C_{d,w}\right) + J_W C_{d,b}}\right] \tag{27}$$

With the above models, given operating condition and molarity of feed and draw solution, iteration procedures are used to predict J_W across the model by Tan and Ng [33]. Tan and Ng [71] described a model for the ECP effect using the boundary layer concept. Qin et al. [72] optimized operating conditions of FO for the membrane bioreactor by conducting experiments at FO pilot plant levels where tap water was considered as feed and NaCl and $MgSO_4$ as draw solutions. Flux under various operating conditions has also been reported [45]. Choi et al. [73] demonstrated FO with osmotic pressure gradient allowing low energy consumption to produce water under ICP. They explained the characteristics of FO and RO processes by applying film theory model for the effect of concentration polarization in FO and RO processes. The flux of combined system using FO and RO was:

$$J_W = L_v\left[\Delta P + \pi_{d,b}\exp\left[\frac{-J_W}{K_D}\right] - \pi_{f,b}\exp\left[\frac{J_W}{K_F}\right]\right] \tag{28}$$

where K_D is the mass transfer coefficient for ICP:

$$K_D = \frac{D\varepsilon}{\tau l} \tag{29}$$

$$K_F = 1.85\frac{D}{d_h^{0.67}L^{0.33}}\left(\mathrm{Re}\,Se\right)^{0.33} \tag{30}$$

This model predicts the internal and ECP effects in FO process and was validated with the lab-scale FO device. Under different operating condition, the calculated flux was compared with experimental flux, and also it was found that the combination of FO and RO may result in a higher flux than FO-only process under some operating conditions by adjusting the back pressure valve. The reverse permeation of draw solution across an asymmetric membrane in FO operation was developed and is validated experimentally with a highly soluble salt (NaCl) as a draw solution [21]. A CA membrane designed for FO and is employed to find membrane transport coefficients, reverse flux selectivity, and the ratio of the forward water flux to the reverse solute between the model predictions and experimental results. A non-invasive direct microscopic observation method for characterizing fouling, draw solution concentration, membrane orientation and feed spacer of an FO membrane was applied by Wang et al. [74] in a cross-flow setup using latex particles as model foulant in the FW. The coupled effect of dilutive internal concentration polarisation (DICP) was demonstrated by Qin et al. [75] to show that concentrative external concentration polarisation (CECP) limits flux of FO. Individual contributions of various types of DICP and CECP were modeled and validated by pilot studies.

Each membrane's hydraulic permeability coefficient (A) and the salt permeability(B) was compared with predicted flux at system operating conditions using mass transport boundary layer theory in a cross-flow FO system [27]. An FO cargo transfer bag (FO-CTB) was developed using a logistics reduction technology by Flynn et al. [76] that can be reused to provide radiation shielding and water recycling capacity on orbit. The effect of microgravity on the FO membrane bags was exploited in the design, construction and testing of a prototype FO-CTB. The CTB was integrated with the habitat demonstration unit to explain basic function of a water treatment system. In a facility with an osmotic membrane bioreactor (OMBR), Holloway et al. [77] demonstrated its superior rejection capability toward dissolved constitution compared with conventional (UFO–MBR) system. The mass balance model for ultrafiltration-osmotic membrane bioreactor (UFO–MBR) process was solved through iterative technique to predict the salt and phosphorous concentration in the anoxic tank [C_{an}] and aerobic tank [C_{FO}] using the following equations:

$$C_{\mathrm{an}} = \left[F_{\mathrm{in}} * C_{\mathrm{in}} + F_{\mathrm{r}} * C_{\mathrm{FO}\,(t_i-1)} - \left(F_{\mathrm{in}} + F_{\mathrm{r}}\right) * C_{\mathrm{an}\,(t_i-1)}\right] * \frac{\left(t - t_{\mathrm{i}-1}\right)}{V_{\mathrm{an}}} + C_{\mathrm{an}\,(t_i-1)} \tag{31}$$

$$C_{\mathrm{FO}} = \left[\left(F_{\mathrm{in}} + F_{\mathrm{r}}\right) * C_{\mathrm{an}(t_i-1)} + J_{\mathrm{rfs}} - \left(F_r + F_{\mathrm{uf}} + F_w\right) * C_{\mathrm{FO}(t_i-1)}\right] * \frac{\left(t - t_{i-1}\right)}{V_{\mathrm{FO}}} \tag{32}$$

where F_{in} is the sum of fluxes of influent water in FO, and F_{uf} is the permeate flow rate in fixed UF. The inputs to the model are measured average TDS concentration (C_{in}) of influent stream and calculated reverse salt flux through the FO membrane (J_{RFS}). C_{an} and C_{FO} were estimated at every 6 h interval using parameters at previous time step (t_{i-1}) under constant recycle flow rate F_r, anoxic tank volume V_{an} and aerobic tank volume V_{ar}. A microbial osmotic fuel cell with FO membrane placed between the electrodes was constructed by Werner et al. [78] who compared the energy recovery and the performance of the cell with the conventional microbial fuel cell fitted with cationic or anionic exchange membranes. Kong et al. [79] investigated that the rejection of haloacetic acids (H_{AA}s) by FO in a coupled reverse DS was mathematically modeled using the solution–diffusion model for both the active layer facing the FW (AL-FW) and the active layer facing the draw solution (AL-DS). The change in weight as function of time was utilized to determine the flux of permeate water:

$$J_w = \frac{\Delta m}{\Delta t A_m} \tag{33}$$

where $\Delta m/\Delta t$ is the rate of change of weight, and A_m is the area of the membrane. The variation of the concentration of H_{AA} in the DS was used to determine the flux of H_{AA} through the FO membrane. The variation of concentration of NaCl in FW was used to determine the flux of reverse solute through the membrane using:

$$J_w = \frac{\Delta(V_c)}{\Delta t A_m} \tag{34}$$

where the $\Delta(V_c)$ is the variation of the target compound in the FW or DS tank during the specified time interval. With the permeate water flux (J_w) and the H_{AA} fluxes (J_{HAA}), the rejection ratio of each H_{AA} compound (R_{HAA}) in the FO process can be obtained from:

$$R_{\text{H}_{\text{AA}}} = 1 - \frac{J_{\text{H}_{\text{AA}}}}{J_m C_{\text{FHAA}}} \tag{35}$$

where C_{FHAA} is the H_{AA} concentration in the FW. For the AL-FW orientation the permeate water flux J_w is expressed as:

$$J_W = A\left[\pi_{D,\text{Nacl}} \exp\left(\frac{-J_W}{K_m,\text{Nacl}}\right) - \pi_{F,\text{Nacl}} \exp\left(\frac{J_W}{K_m,\text{Nacl}}\right)\right] \tag{36}$$

For AL-FW orientation case, the reverse DS flux J_{NaCl} can be expressed as:

$$J_{\text{Nacl}} = B_{\text{Nacl}}\left[C_{D,\text{Nacl}} \exp\left(\frac{-J_W}{K_m,\text{Nacl}}\right) - C_{F,\text{Nacl}} \exp\left(\frac{J_W}{K_m,\text{Nacl}}\right)\right] \tag{37}$$

The flux of H_{AA} through the FO membrane can be expressed as:

$$J_{\text{H}_{\text{AA}}} = \frac{B_{\text{H}_{\text{AA}}} \exp\left(\frac{J_W}{K_m,\text{H}_{\text{AA}}}\right)}{1 + \frac{B_{\text{H}_{\text{AA}}}}{J_W}} C_{F,\text{H}_{\text{AA}}} \tag{38}$$

where A and B are the water permeability coefficient and solute (DS or H_{AA}) permeability coefficient. K_m is the mass transfer coefficient:

$$K_m = D/S \tag{39}$$

where S parameter is the intrinsic length scale of ICP in the FO membrane support layer. $S = (t \times \tau/\varepsilon)$ where t is the thickness; τ is the tortuosity; and ε is the porosity of the support layer.

For the AL-DS orientation case, the permeate water flux J_w is expressed as:

$$J_W = A\left[\pi_{D,\text{Nacl}} \exp\left(\frac{-J_W}{K_m,\text{Nacl}}\right) - \pi_{F,\text{Nacl}} \exp\left(\frac{J_W}{K_m,\text{Nacl}}\right)\right] \tag{40}$$

For AL-DS orientation case, the reverse DS flux J_{NaCl} can be expressed as:

$$J_{\text{Nacl}} = B_{\text{Nacl}}\left[C_{D,\text{Nacl}} \exp\left(\frac{-J_W}{K_m,\text{Nacl}}\right) - C_{F,\text{Nacl}} \exp\left(\frac{J_W}{K_m,\text{Nacl}}\right)\right] \tag{41}$$

The flux of H_{AA} from the FW to DS can be expressed as:

$$J_{\text{H}_{\text{AA}}} = \frac{B_{\text{H}_{\text{AA}}} \exp\left(\frac{J_W}{K_m,\text{H}_{\text{AA}}}\right)}{1 + \frac{B_{\text{H}_{\text{AA}}} \exp\left(\frac{J_W}{K_m,\text{H}_{\text{AA}}}\right) \exp\left(\frac{-J_W}{K_m,\text{H}_{\text{AA}}}\right)}{J_W}} C_{F,}\text{H}_{\text{AA}} \tag{42}$$

The NaCl permeability coefficient, B_{NaCl}, was derived from the rejection ratio vs. applied pressure by filtering NaCl solution:

$$R = \frac{1}{1 + \frac{B}{A(\Delta P - \Delta\pi)}} \tag{43}$$

Lee et al. [67] presented fouling behavior along the length of membrane module by providing simple modeling and laboratory-scale experiments of FO membrane process. This model reveals that, with decrease in differential osmotic pressure across the FO membrane, the permeate flux decreases along the membrane channel. McGinnis and Elimelech [65] and McGinnis et al. [66] used Hyprotech systems (HYSIS) to calculate energy requirements of ammonia–carbon dioxide in FO desalination. The FO process model, assumed to be combination of single/multiple separation columns engaged in separating DSs from the product water was used to calculate the thermal and electrical energy balances for the process. These were compared with the energy requirements of current desalination methods. A 2D numerical model representing coupled hydrodynamics and mass transfer equations was formulated for FO desalination [1] and was solved for cases where: (i) the membrane skin faces the salt feed solution and (ii) the membrane skin faces the draw solution. Kumar and Pal [80] designed FO–NF integrated system in flat-sheet cross-flow module to separate reusable water from coke-oven wastewater with reduced concentration polarization and high flux using low energy. Deshmukh et al. [82] developed a model for estimating the volumetric energy efficiency of PRO via pressure–volume analysis of a membrane process and requires only the osmotic pressure, π, and mass fraction, w, of water in the concentrated and dilute feed solutions to estimate the maximum achievable specific energy density, u, as a function of operating pressure. Is independent of any membrane or module properties. It is possible to examine the effects of volumetric solution cost, operation of a module at various pressure, and operation of a constant pressure module with various feed. Wendt et al. [81] and Welty et al. [70] formulated a model to calculate energy requirements of a switchable polarity solvent FO (SPS-FO) plant for purification of water from aqueous NaCl feed and claimed that the technology is at par with the cost of seawater desalination by RO. Fig. 3 shows block diagram of SPS-FO process model [70].

Deshmukh et al. [82] formulated a scale model to analyze the effects of membrane's water permeability, solute permeability and its structural parameters, on the overall performance of an FO plant to obtain water from seawater. The model was simulated with 0.6 M NaCl under the thermodynamic limit of operation. The results indicated that the maximum achievable water recovery is practically independent of membrane properties, while higher maximum water recovery is possible in a counter-current setup compared with co-current mode [37]. The transport/separation of boron through FO membranes can be predicted to guide the fabrication of improved membranes with decreased permeability of boron and structural parameter to minimize flux of boron. Both theoretical model and experimental results demonstrated that flux (boron) was substantially greater when membrane active layer was facing draw solution, compared with the other membrane orientation due to more severe ICP and the rejection of contaminants was found in FO processes by Jin et al. [10]. Double-skinned FO membranes were designed for boron rejection up to 83.9% for boron permeation studies, which was demonstrated by theoretical calculations, followed by experiments. The double-skinned FO membrane was fabricated using a sulfonated polyphenylenesulfone polymer as the hydrophilic substrate and polyamide as the selective layer material via interfacial polymerization on top and bottom surfaces. The effects of key parameters on rejection of boron, such as its permeability through both selective layers and structural parameters, were also investigated and not only provide insight for boron removal from wastewater but also showed enlightenment toward the design of next generation FO membranes to eliminate low-rejection molecules in wider applications by Luo et al. [11].

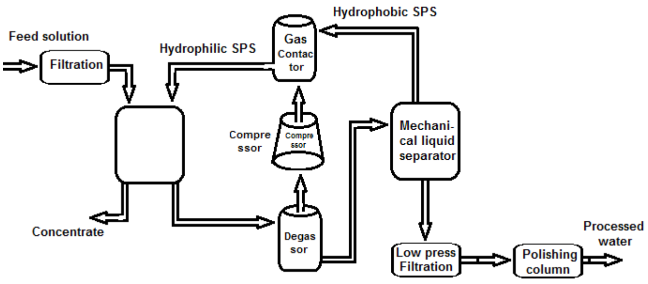

Fig. 3. The block diagram representation of SPS-FO process model.

5. Identification of parameters for FO desalination process

After validating models of FO, it is needed to identify model parameters of process (for synthesizing a low-order model structure) for designing closed-loop control of process variables (throughput demand of permeate and its parts per million (ppm) level or conductivity) by online monitoring and adjusting input variables (feed pressure of pump and pH of feed). The performance of closed-loop system depends on perfect controller tuning, which needs a model-based tuning rule, a process. As the model is perfectly validated, it can be used for tuning of controller parameters. The controller tuning is done by applying identification or estimation of transfer function technique. Many researchers have suggested identified transfer functions from input/output data of FO process. FO is a solution osmotic pressure process, used for removing low molecular weight solutes, such as inorganic salts or small organic molecules from a solvent. FO was used in the field of food processing by Lee et al. [83] who observed that the performance of this asymmetric membrane is poor due to boundary layer that occurs within the supporting layer for osmotic-driven pressure.

The feed is the seawater, and the draw solution is the ammonium carbonate (or) sucrose as suggested by Low [27]. The block diagram of FO system with manipulated variables, disturbance variable and controlled variables are shown in Fig. 4. An additional distinction between the RO and FO processes is that the water permeating the RO process is in most cases freshwater ready for use. In the FO process, this is not the case. The membrane separation of the FO process in effect results in a "trade" between the solutes of the feed solution and the draw solution.

Lee et al. [83] suggested integral equations to analyze non-linear behavior of osmotic pressure of solution consisting of high concentration of inorganic salt. Interaction between the molecules is assumed to be Lennard-Jones (LJ) potential, and various force fields were used to determine potential parameters. Relationship between LJ parameters and permeate flux are discussed.

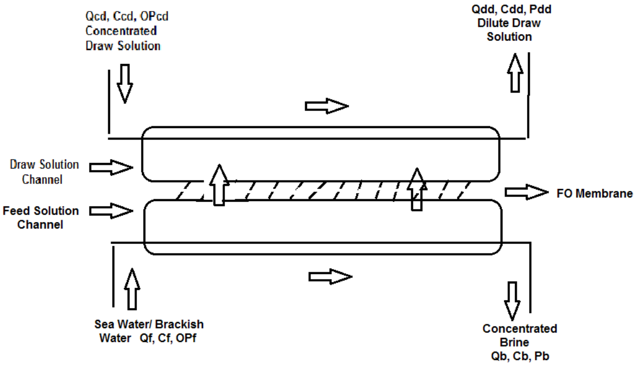

Fig. 4. Block diagram of FO system with manipulated variables, disturbance variable and controlled variables.

Few studies have been reported to make hybrid of FO and MDs for increasing the throughput of the process. These process technologies described above are used in different applications. Following section describes the developments in this regard.

6. Application of FO technique and hybrid processes

Salter [84] used hydration process with an ingestible DS (sugars, glucose or fructose) to separate water from dilute feeds containing pathogens or toxins that were readily rejected by the membrane. The technology was used to recycle urine (of a backpacker or soldier to survive) in arid environments. Highly concentrated saline FW was used to produce potable water from an FO system with ingestible solutes [51]. This may find an application for survival in life rafts at sea. A second separation step is required when the desired product is freshwater, which does not contain DSs (Fig. 3). Here, the first separation step of FO, driven by an osmotic pressure gradient, does not require a significant energy input (only unpressurized stirring or pumping of the solutions involved). The second separation step, however, does typically require energy input. One method used for the second separation step is to employ RO. This approach has been used, for instance, in the treatment of landfill leachate. An FO membrane separation is used to draw water from the leachate fed into saline (NaCl) brine. The diluted brine is then passed through an RO process to produce freshwater and a reusable brine concentrate. The advantage of this method is not energy savings, but rather in the fact that the FO process is more resistant to fouling from the leachate feed than an RO process alone would be [85]. A novel NF hollow fiber membrane made of PBI has been developed as an FO membrane. A similar FO/RO hybrid has been used for the concentration of food products, such as fruit juice [86].

Cath et al. [87] presented the impacts of brine mismanagement and have drawn attention to new methods for enhancement of water recovery in desalination processes using membrane contactor processes, namely where FO and MD are tested. Pressure-driven membrane processes (e.g., RO and NF) are limited to moderate recoveries by scale formation and require intensive chemical and physical treatment for higher recoveries. The thermal processes can achieve higher recoveries but are energy intensive. This method is found to be efficient compared with other methods [88] investigated that FO is a membrane treatment process at bench scale to both batch and continuous operating conditions. The continuous bench-scale system used FO as pre-treatment for RO, which will yield high water flux and high nutrient rejection, and they developed the mathematical model to determine the specific energy, power and membrane area requirements for a larger-scale centrate treatment process. Tan and Ng [71] suggested that the FO process can be used as an alternative to traditional brine treatment technologies due to its low energy requirement. Choi et al. [73] suggested that FO enables lower energy consumption to produce water than RO because it does not require high transmembrane pressure and FO needs to be combined with other processes such as RO or evaporation because the draw salts in the FO products should be removed. In their article, they focused on theoretical investigation of combined systems with FO and RO for seawater desalination. A theoretical model [30,49] to predict the performance of the combined systems was developed based on the solution diffusion model. Brian et al. [9] completed a review on the literature on MD and FO for treating natural and recovered wastewaters. As alternatives to pressure-driven processes such as RO, which are expensive in both capital and energy, require pre-treatment of the FW, MD with hydrophobic microfiltration membranes that can make use of low-grade heat energy, to give higher yields of product water from concentrated FWs. FO uses hydrophilic membranes akin to RO. Tan and Ng [71] proposed the hybrid FO–NF process for seawater desalination with seven potential draw solutions. The FO–NF process was investigated using laboratory-scale FO and NF test cells. Results from both FO and NF tests suggested that the hybrid FO–NF process is a feasible process for seawater desalination. Water fluxes of about 10 L/m^2 h, for both FO and NF processes could be achieved. Solute rejection of the FO membrane was maintained at over 99.4% for all seven DSs tested. Mi et al. [60] investigated the role of various physical and chemical interactions, such as intermolecular adhesion forces, calcium binding, initial permeate flux, and membrane orientation, inorganic fouling of FO membranes. Alginate, bovine serum albumin (BSA) and Aldrich humic acid were chosen as model organic foulants and quantified the intermolecular adhesion forces between the foulant using atomic force microscopy. Cornelissen et al. [54] developed an innovative OMBR, based on FO driven by an osmotic pressure difference. For the OMBR to be both technically and economically viable, the performance of the FO membranes has to be sufficiently high, while membrane fouling and draw solution leakage have to be low. The effect of temperature, membrane type, membrane orientation and concentration of draw solution was studied to optimize FO membrane performance using DI water and membrane fouling, and draw solution leakage was studied in this work by laboratory-scale FO experiments using an activated sludge. Xu et al. [55] suggested that the FO process potentially can be used as an alternative to traditional brine treatment technologies because of its low energy requirement and the feasibility of concentration of brine by the FO process was studied using a laboratory-scale unit. An FO membrane and the dense selective layers (i.e., with their support layers properly peeled off) of two types of RO membrane (i.e., cellulose acetate (CA) and polyamide composite, denoted as CA and amide dianhydride (AD), respectively) were tested where it was found that the dense selective layer of the CA membrane could achieve the highest water flux. Hancock and Cath [89] explored the different operating condition on the forward diffusion of solute found in brackish water and seawater and reverse diffusion of common draw. Bamaga et al. [90] introduced FO-driven process that has been used for membrane filtration (MF), UF and NF membrane pre-treatment instead of conventional pre-treatment in sea water reverse osmosis (SWRO) process. Martinetti et al. [91] studied vacuum-enhanced direct contact membrane distillation (VEDCMD) and FO for water recovery enhancement in desalination of brackish water. High water recoveries were obtained using two RO brine streams with total dissolved solids concentrations averaging 7,500 and 17,500 mg/L that were further desalinated by VEDCMD and by FO, and these recoveries were limited by precipitation of inorganic salts on the membrane surface. Achilli et al. [92] presented a novel OMBR, which utilizes a submerged FO membrane module inside a bioreactor through which water is transported from the mixed liquor across a semi-permeable membrane, and into a draw solution (DS) with a higher osmotic pressure. Potable water is produced by treating the dream-initiated lucid drinking DS in an RO unit, and the by-product is a reconcentrate. DS stream is for reuse in the FO process. From the experiments, it was demonstrated that the flat-sheet cellulose triacetate FO membrane has high sustainable flux and relatively low reverse flow. McCormick et al. [93] reported the diffusive permeance of water, NaCl and ethanol through several, unoptimized membranes, and these reports can facilitate analytical and development of water recovery from highly impaired sources using hybrid processes based on forward (direct) osmosis (FO) with aqueous ethanol solutions as the osmotic agent. Studied membranes included anion and cation exchange materials and cross-linked polyvinyl alcohol gels, the latter being a membrane chemical commercially used for ethanol dehydration via par-evaporation and measured transport properties.

According to Hau et al. [46] the major challenges of FO technologies are: (i) ineffective membranes that are heart of most FO-based processes; (ii) lack of cost-effective DSs that can be easily recycled and (iii) limited studies on fouling. For direct fertigation and osmotic power generation, fertilizer and seawater were natural DSs [69]. However, osmotic power generation can be carried out by using PRO, which is not the theme of this article. Molecular design of FO membranes has high flux and power density. Conventional FO membranes are designed for no-pressure or low-pressure operation environments. Recommended FO membranes are likely to be damaged under this high pressure condition. Coday et al. [8] found that many traces of organic compounds including pharmaceuticals, health care product, disinfection by-product and industrial chemicals cause serious concern that need to be addressed for preventing contamination of water. They suggested removal of these compounds by FO process. Table 2 highlights benefits of FO in different areas or fields of application.

Tian et al. [50] reported that novel OMBRs use a dense salt-rejecting FO membrane and exhibit high retention of organic matter and various other contaminants. Hickenbottom et al. [94] presented the extract of shale gas and coal methane gas hydraulic fracturing of wells, and novel application of FO was experimented for treatment and reclamation of water from drilling waste to give water reuse. Table 2 highlights benefits of FO in different areas or fields of application. El-Saie et al. [95] compared the performances of RO, FO–RO and FO processes as summarized in Table 3. From the above discussions, the development of FO systems and their improvement are clear. Now, we need to analyze what are the benefits of FO system compared with conventional RO systems. The following section will describe the advantages of an FO process. It can be observed that FO requires less energy to produce per cubic meter of water than the other thermal and membrane techniques. The cost required to construct an FO plant is considerably less than other thermal technologies, even RO. For 1 million gallons per day (mgd) plant of RO may cost 9.96 million USD for construction and 1.06 million USD/year for operation. Whereas, construction cost of FO plant (same capacity) may be 8.78 million USD needing an operating cost of 0.83 million USD/year. A plant operating on FO technology can be constructed at 90% of the construction cost and operated at 80% of the operation cost of an SWRO plant, with the current options of FO membranes and draw solution available. A typical hybrid system of FO–RO can reduce the operating cost up to 45%. Hancock and Cath [89] estimated 0.43 USD/m^3 cost savings using FO–RO hybrid system compared with stand-alone RO. They suggested a capacity of 30 L/m^2 h as minimum average permeation flux to run an FO with economic sustainability. Compared with RO, FO has less membrane fouling, scaling and brine discharge.

In order to improve the flux some studies have been carried out by combining FO and MD techniques, which are discussed as follows:

Nayak and Rastogi [96] compared the osmotic MD and FO membrane processes for concentration of anthocyanin extract to study the effect of various process parameters such as osmotic agent concentration, flow rates of feed, osmotic agent on transmembrane flux and mass transfer in case of osmotic

Table 2
Benefits and challenges of different applications of FO

Applications of FO	Benefits	Challenges
Desalination	Low energy consumption for water transport across the semi-permeable membrane	Ineffective membranes; lack of cost-effective draw solutes
Direct fertigation	Fertilizers are natural draw solutes; diluted draw solution is useful for irrigation	Limited application sites
Osmotic power generation	Seawater is a natural draw solute	Pre-treatments of seawater and river water; complicated fouling phenomenon owing to the high pressure in the seawater compartment. Osmotic membrane bioreactor low fouling and low energy consumption need to find low cost and easy recyclable draw solutes. However, osmotic power generation can be better carried out by using pressure retarded osmosis
Osmotic membrane bioreactor	Low fouling and low energy consumption	

Table 3
Comparison of the performance of the RO, FO–RO and FO processes

S. No.	Parameters	RO	FO–RO	FO
1.	Water flux	High	Low	Low
2.	Operating cost and power consumption	High (decreases for high water salinity) 3.5 kWh/m^3	Decreases for high water salinity	Small (2%–4% of power consumption by FO–RO)
3.	Efficiency		Higher for higher seawater salinity	
4.	Draw solution regeneration	Best		
5.	Permeate TDS		Increases with the increase in concentration of draw solutes (the lowest permeate TDS was achieved in the 0.65 mol $MgCl_2$ FO–RO process followed by the RO process, 1 mol NaCl FO–RO and 1.2 mol NaCl FO–RO processes, respectively)	
6.	Reliability	High		
7.	Maximum water recovery rate	High	Increases with the increase in the concentration of draw solutes	
8.	Salt rejection rate	High	Low	Very low
9.	Treat wide range of seawater concentration	Best		
10.	High pressure pump	Needed	Needed	No need

MD and FO. Xie et al. [97] presented simultaneous separation of phosphorus and clean water from digested sludge concentrate using an FO – coupled with a MD. In this hybrid process, FO–MD first concentrates orthophosphate and ammonium for subsequent phosphorus recovery in the form of struvite ($MgNH_4PO_4.6H_2O$), while MD recovers draw solution and separates clean water from the digested sludge. Qin et al. [15] and Wang et al. [34] were able to discriminate individual contribution of different types of DICP and CECP via modeling and validated it by pilot studies. The influence of DICP/CECP on FO flux has been investigated in this study. A comparison between MD and FO–MD processes has been carried out in Table 4. Table 4 summarizes differences between MD and FO–MD processes from direct sewer mining. From the above discussions, the development of FO systems and their improvement are clear. Now, we need to analyze what are the benefits of FO system compared with conventional RO systems. The following section will describe the advantages of an FO process.

Zou et al. [43] reported that novel OMBRs uses a dense salt-rejecting FO membrane and exhibits high retention of organic matter and various other contaminants. Hickenbottom et al. [94] presented that the extract of shale gas and coal methane gas hydraulic fracturing of wells and novel application of FO was experimented for treatment and reclamation of water from drilling waste to give water reuse. OMBR is used to treat wastewater. A mathematical model for full-scale OMBR was formulated to simulate salt accumulation, draw solute (DS) dilution and flow of water flux through hollow fiber membrane of OMBR using design parameters, properties of DS and characteristics of FO membrane. The results showed decline of water flux from 10 to 0.82 L/m^2 h with reduction in retention time of smaller solids to solid retention time (SRT): 5 d due to salt accumulation and DS dilution. When OMBR is scaled up using commercially available DS and FO membrane, it resulted into reverse salt flux, ICP and ECP. It was observed that ECP adversely impacts water flux in full-scale OMBR although it was often ignored in previous works by Kim [98]. Park et al. [99] developed a full-scale model of OMBR to find optimal design parameters OMBR-RO hybrid system for wastewater reclamation. It was simulated to find salt accumulation, draw solution dilution and water flux that considered sludge concentrator for high retention and low salt concentration factor [99]. In order to predict salt accumulation and draw solution, Kim [98] proposed a model for OMBR using hollow fiber membrane and found that growth of microbes is affected by concentration of salt.

7. Comparison of RO and FO

Tan and Ng [71] compared the performance of a forward (direct) osmosis (FO) process using a laboratory-scale unit to expose the effect of membrane structure and orientation on water flux. Nayak and Rastogi [96] compared the FO desalination and conventional RO, from both real operational results and theoretical comparison looking at the merits and demerits of the respective processes. This is of particular relevance when more complex pre- and post-treatment such as MF or UF and boron removal are applied to RO. Energy consumption figures with respect to the degree of fouling are presented.

Hau et al. [46] done the comparison of the fouling behaviors during FO and RO. Alginate, humic acid and BSA are used as organic foulants, and two suspensions of silica colloids of different sizes are chosen as model particulate foulants.

Table 4
Comparison of MD process with FO–MD process based on parameters

S. No.	Parameters	MD process	FO–MD process
1.	Membrane	Hydrophobic, microporous membrane for MD process	Flat sheet cellulose membrane for FO process; Hydrophobic, microporous membrane for MD process
2.	Permeate flux for cumulative permeate volume of 4,000 mL	4 L/m^{2}h	7 L/m^{2}h
3.	Permeate flux for clean feed for cumulative volume of 4,000 mL	–	8 L/m^{2}h
4.	Permeate flux for raw feed for cumulative volume of 4,000 mL	–	6.5 L/m^{2}h
5.	% Rejection for cumulative volume of 4,000 mL	96%	100%
6.	Permeate conductivity in µS/cm for cumulative volume of 4,000 mL	3	9
7.	Rejection of trace organic compounds		
8.	(a) Pentachlorophenol(n)	70%	94%
	(b) Triclosan(n)	84%	99%
	(c) Linuron(n)	85%	94%
	(d) Atrazinc(n)	90%	98%
	(e) Amitriptyline(+)	99%	99%
	(f) Diclofenac	100%	99%
	(g) Carbamazepine(n)	99%	99%
	(h) Sulfamethoxazole	99%	99%
	(i) Caffeine(n)	99%	99%
	(j) Trimethoprim(+)	100%	99%
	(k) Primidone(n)	98%	99%
	(l) Bezafibrate	100%	100%
9.	For feed concentration of 6,000 ng/L	Product 1,500 ng/L	Draw concentration of 3,000 mg/L and product concentration of 1,500 mg/L
10.	Recovery rate	Low	
11.	Thermal efficiency	High	
12.	Power consumption	Low	

Identical hydrodynamic operating conditions (i.e., initial permeate flux and cross-flow velocity) and FW chemistries (i.e., pH, ionic strength and calcium concentration) are employed during FO and RO fouling runs. The flux-decline behavior in FO changed dramatically with the type of organic foulant, size of colloidal foulant and the type of the draw solution employed to generate the osmotic driving force. In FO, reverse diffusion of salt from the draw solution to the feed side exacerbates the cake-enhanced osmotic pressure within the fouling layer. The elevated osmotic pressure near the membrane surface on the feed side leads to a substantial drop in the net osmotic driving force and, thus, significant decline of permeate flux. The structure (i.e., thickness and compactness) of the fouling layers of FO and RO is quite different. By varying the cross-flow velocity during the organic fouling runs, fouling reversibility in FO and RO are examined. The permeate flux during organic fouling in FO recovered almost completely with increasing cross-flow velocity, while no noticeable change was observed for the RO system. Organic fouling in FO could be controlled effectively by optimizing the hydrodynamics in the feed stream without employing chemical cleaning. Achilli et al. [14] suggested that RO or a distillation process can be utilized to reconcentrate the DS for reuse in the FO process and to produce purified water. The main advantages of FO include operation at very low hydraulic pressures, high rejection of brine [17]. They also described a schematic for the new hybrid osmotic dilution process using FO membrane [16]. In the first osmotic dilution stage, seawater is diluted by an impaired water stream which is then processed through an RO desalination system, which rejects salts and dissolved contaminants that may have crossed the membrane from the impaired water source. The discharge is further concentrated in the second stage to reduce the volume of the impaired water stream. During dilution, the energy required for RO desalination is reduced. Thus not only the energy but also the rejected contaminants are reduced. Nicoll [68] presented some aspects of FO coupled with RO using a recirculating draw solution, to produce desalinated water for dewatering/concentrating a feed stream. FO process offers reduced rates of fouling to operate the RO step at optimum conditions. McGovern and Lienhard [100] gave a comparison of the theoretical and actual energy requirements of FO and RO seawater desalination and the RO is significantly more energy efficient and that FO research efforts would best be fully oriented toward alternate applications due to draw-dilution step, which increases the theoretical and actual energy requirements for draw regeneration. The regeneration process must be significantly more efficient than RO by optimization of the draw solution and the benefits of reduced fouling during regeneration and to increase the efficiency of an optimal draw regeneration process.

8. Control aspects

Automatic control of FO process comes as second step after development of process design and has been researched by very less authors. Basically, the process variables (manipulated) are osmotic pressure and ratio of feed to recycled stream of reject. There are many control variable, such as, permeate concentration, pH and flow-rate. Once the operating conditions are optimized, the next task is to implement control loops for safe operation and control of the process. Optimization of desalination using FO was discussed by. Moody and Kessler [30]. Brine concentration control for FO was discussed by Yong-Ng and Wanling [101]. FO has been applied to various water treatment problems because of low energy consumption and lower fouling properties [102]. To resolve biofouling issues phosphate limitation was investigated to reduce befouling instead of commonly used chemical cleaning. They maintained FO performance by limiting phosphate in FW that reduces microbial growth and biofilm formation. An accelerated biofilm-enhanced osmotic pressure device was considered to explain control of biofouling through limitation of phosphate in feed using Gibbs–Donnan equilibrium mechanism [102]. *Pseudomonas aeruginosa* PAO1 was selected as model bacterium to verify the effects of phosphate limitation non-biofilm formation in wastewater. A difference in concentration between feed and draw solution led to flux of water that gradually enriched feed solution, and dilute the draw solution. The acquired water and reverse salt flux was used as control data to correct and compare data gained from the fouling experiments. Wang et al. [74] proposed compensation techniques for water loss to maintain proper cooling water quality. Freshwater (rain water as make up water) was added to the circulating system to maintain proper temperatures for different industrial processes. Average water flux was 1.75 $L/m^2{\cdot}h$ at 23°C decreased gradually to 0.65 $L/(m^2{\cdot}h)$ after the draw solution was diluted 4 times. However, an increase in temperature of draw solution from 3°C to 50°C, improved membrane fluxes up to 10 times. Solute losses were made up by adding Na_2HPO_4 to the cooling water periodically. The effects of the water temperature on the water flux were due to osmotic pressure, physio-chemical properties of the solution and the structural parameters of the FO membrane. They suggested running the FO system continuously with nominal water flux to solve issues like high draw solution temperature while getting low membrane fouling rate. They claimed that FO rainwater treatment will provide a potential technology for cooling water dilution and reuse. Linares et al. [103] developed the FO membrane (TFC) technology to remove boron present in seawater using lesser energy. The effect of permeate flux, membrane structure, DS charge and reverse solute flux on boron removal were reported. They claimed 40% higher boron rejection and 20% higher permeate flux compared with conventional procedure.

9. Future direction of research

One area of current research in FO involves the direct removal of DSs by thermal means with effective utilization of energy. This process is typically referred to as the "ammonia–carbon dioxide" FO process, as the DSs are salts formed from the mixing of ammonia and carbon dioxide gases in water [20]. These salts can reach up to high concentrations, particularly as the ratio of ammonia to carbon dioxide is increased. An especially convenient property of these salts is that they readily dissociate into ammonia and carbon dioxide gases again, if a solution containing them is heated (to ~60°C, at 1 atm pressure). Once the concentrated draw solution is used to effect separation of water from the FO feed solution, the diluted draw solution is directed to a reboiled stripper (distillation column) and the solutes are completely removed and recycled for reuse in the FO system [14]. An FO system of this type thereby effects membrane separation of water from the FO feed, using heat as its primary energy source. The quality of heat used by this process can be very low, at temperatures as low as 40°C. If FO of this type is used in a cogeneration environment (e.g., waste heat from a power plant), its energy cost can be greatly reduced compared with RO [55,56].

A second area of current research in FO also involves direct removal of DSs, in this case by means of a magnetic field. Small (nano-scale) magnetic particles are suspended in solution creating osmotic pressures sufficient for the separation of water from a dilute feed. Once the draw solution containing these particles has been diluted by the FO water flux, they may be separated from that solution by use of a magnet (either against the side of a hydration bag, or around a pipe in-line in a steady-state process). Third, research on (FO–MD) hybrid FO may consume less total energy for operation when thermolytic solutions are used as DS. Hybrid FO–RO may reduce scaling and fouling thereby can achieve higher water recovery at a higher operating pressure. Further optimization on operating parameters, membrane permeability, rejection and packing density may allow use of NF–RO type hybrid systems. Introduction of smart and innovative DSs (MNPs, stimuli-responsive polymer hydrogels) with long life may improve the functioning of FO systems

10. Conclusion

This paper reviews technologies of desalination and separation using FO. Osmotic pressure gradient is the driving force for the separation of solvent from the dissolved solute in FO using semi-permeable membrane. Mathematical modeling is essential to study optimization, scale up for commercialization etc. A quite number of papers reviewed experiments and formulated steady-state/transient mathematical models. These models can be arranged into linear form for designing controllers. They mostly highlighted flux, membrane properties, fouling and applications. Properties of DS are also important criteria in designing the FO process. Process control techniques will help in making operational strategy through a safer one. Hence, detailed reviews on mathematical modeling, identification and control of FO process have been discussed here using 105 papers in related fields. Some highlights on future direction of research have been thrown to motivate further research in this area. Upscaling of units should be done incorporating fouling models into it. Though there is a need to improve flux in FO system, consideration on increased fouling, lower rejection is a concern. However, addition of antiscalant can be advised. This review discusses development of membrane, concentration polarization, novel membranes, and selection of DS, hybrid systems, identification and control aspects for safe operation of FO. Though FO–RO schemes require further validation, it promises radical shift in current consideration of water purification. It is also questionable whether FO/PRO–RO hybrid process will allow sustainable technology with higher flux rate or not. Pressure assisted FO–RO system may be more efficient than RO system for seawater at 40 g/L. This state-of-art review will help researchers to understand the available green technologies, means of their improvement and strategies of safe operation and control through implementation of model-based control techniques.

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