Fouling in Forward Osmosis Membranes: Mechanisms, Control, and Challenges

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Additional information is available at the end of the chapter

Abstract

Continuously escalating global water demand places a substantial burden on the available water and energy resources. Forward osmosis (FO) is an evolving membrane desalination technology that has recently raised interest as a promising low-energy process. FO is a unique method since it utilizes natural osmosis as the driving force, and hence, it ensures that the energy consumption is significantly reduced, in comparison to other pressure-driven membrane processes that are constrained by their excessive energy consumption and unsustainable cost. Therefore, the growing interest in FO from various disciplines and industrial sectors calls for a better understanding of the FO process and further advances in the FO technology management. This chapter aims to provide an in-depth assessment of the water transport phenomenon in FO membranes by focusing on the influence of internal concentration polarization, membrane structure/material, and membrane orientation on the permeate flux. This chapter offers critical insight that can lead to the potential development of new FO membranes with reduced internal concentration polarization and higher water permeability. In addition, key strategies for FO membrane development, some of its challenges, and the perspectives for future investigations of FO membrane fouling and effective FO fouling control methods are explored in this chapter.

Keywords: forward osmosis, fouling, concentration polarization, mass transfer, water filtration

1. Introduction

As the fossil fuels are depleted and the world population continues to rapidly increase, energy and water became two of the most vital global resources. Energy emergencies and the lack of

water have severely affected communities worldwide [1–3]. Reports indicate that more than 1.2 billion people do not have access to safe and clean drinking water sources, while 2.6 billion do not have adequate levels of sanitation [1, 4, 5]. In fact, the overall annual financial loss in Africa caused by the lack of access to basic sanitation and clean water is valued at $28 billion, or 5% of Africa's gross domestic product [5]. While oceans are covering the majority of the planet's surface, only 0.8% of the world's water can be defined as potable [6]. Moreover, the recent world energy outlook report [2] indicates that the world's marketed energy use is predicted to rise by 49% from 2007 to 2035. Data such as this reflects a dangerous trend, especially since currently 1.5 billion people, or more than 1/5 of the world's population, still do not have access to reliable electricity.

Interdisciplinary research groups need to remain aware of the explicit connection that exists between energy and water. The process of making freshwater accessible is a highly energy-demanding process, while the production of the required power frequently necessitates substantial amounts of water [7, 8]. A relatively new technology, forward osmosis (FO), shows a lot of potential in energy production and water supply, especially for applications in controlled-release–type drug medication, medical product enrichment, and food processing. Over the last decade, FO has incited substantial interest in the areas of seawater/brackish desalination [9–11], food processing [12–15], power generation [16–19], and wastewater treatment [20–22]. In terms of its methodology, FO is an osmotically driven membrane process that relies on the osmotic pressure gradient and that moves water across a semipermeable-type membrane from the feed solution side, with the low osmotic pressure, to the draw solution side, featuring high osmotic pressure. Because of its lower hydraulic pressure demands, FO provides multiple benefits, such as lower fouling tendency, easier fouling removal [20, 22, 23], smaller energy input [24], and greater water recovery [25, 26], if compared to pressure-driven processes such as ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO).

2. Advantages of forward osmosis

There are numerous potential benefits offered by FO, especially because of the lower hydraulic pressure values necessary for this osmotically driven–type process. FO's benefits are reflected by its various water treatment applications. First, FO can help obtain smaller energy consumption potentials and as a consequence lower the overall costs and contribute to the production of technically and economically innovative solutes and their respective regeneration methodologies [3, 18, 24]. Arguably, this is one of the key advantages of FO, considering the ongoing global energy crisis. Research studies have shown that membrane fouling in FO is comparatively small [20], somewhat more reversible [23, 27], and may be lowered using hydrodynamics optimization [28]. Furthermore, a number of contaminants may be successfully filtered out with the aid of the FO process [29, 30]. FO can likewise feature greater water recovery and improved water flux because of the higher osmotic pressure gradient occurring across the membrane. Greater water recovery can help reduce the desalination brine volume, especially as it is a substantial environmental concern when it comes to desalination plants and inland desalination facilities [9]. Moreover, in the industries like pharmaceutical and food processing,

FO offers the benefits of preserving the physical properties of the feed, such as color, aroma, nutrition, and taste, without diminishing the overall quality, as it is not heated or pressurized [14, 31, 32]. When it comes to medical uses, FO can help with the release of drugs featuring low oral bioavailability, or poor solubility, in a controlled way and implementing osmotic pumps [33, 34].

3. Modeling of water transport in forward osmosis

The general equation for water flux in forward osmosis (FO), reverse osmosis (RO), or pressure-retarded osmosis (PRO) is [16]:

$$J_w = A(\sigma \Delta \pi - \Delta P) \tag{1}$$

where J_w is the water flux, A is the membrane's water permeability constant, σ is the reflection coefficient, $\Delta \pi$ is the osmotic pressure difference across the membrane, and ΔP is the applied hydraulic pressure variance. In Eq. (1), the term ($\sigma \Delta \pi - \Delta P$) signifies the effective driving force necessary for the water molecules' transport across the membrane. In the FO desalination process, there is no hydraulic pressure applied and the change in osmotic pressures is the sole driving force; Eq. (1) can be expressed as:

$$J_w = A\sigma \Delta \pi_{Bulk} = A\sigma(\Delta \pi_{Draw} - \Delta \pi_{Feed}) \tag{2}$$

where $\Delta \pi_{Feed}$ stands for the feed solution's bulk osmotic pressure, and $\Delta \pi_{Draw}$ is the draw solution's bulk osmotic pressure. Eq. (2) is restricted by the assumption that the membrane does not permit draw solute permeation [35, 36]. Furthermore, Eq. (2) is applicable for dense symmetric membranes, in which the driving force for water molecules is the difference between the osmotic pressures of the bulk feed and draw solutions, as reflected in **Figure 1**.

If it can be assumed that the difference between the bulk osmotic pressure of the feed and the draw solution is the driving force responsible for water permeation through membranes in FO, then Lee et al. [37] proposed the following model for low water flux cases:

$$J_w = \frac{1}{K} ln\left(\frac{\pi_{Draw}}{\pi_{Feed}}\right) \tag{3}$$

where K stands for the resistance to diffusion of solute within the porous support layer of the FO membrane, and π_{Draw} and π_{Feed} are the respective bulk osmotic pressures of the draw and the feed solution. K can be estimated using Eq. (4) [16]:

$$K = \frac{t\tau}{\varepsilon D} = \frac{S}{D} \tag{4}$$

where t is the membrane's thickness, τ is tortuosity, ε is membrane porosity, S is membrane's structural parameter, and D is the solute's diffusion coefficient.
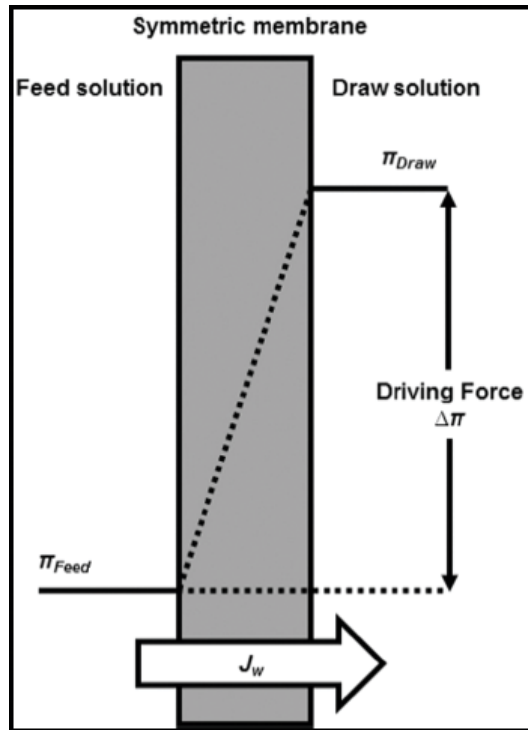
Figure 1. Ideal osmotic pressure driving force in the case of symmetric membranes.

4. Challenges in forward osmosis

FO applications are still facing some critical challenges even though the osmotically driven membrane processes have been extensively researched in relation to a range of applications and environments. FO's primary issues are connected to aspects such as membrane fouling, reverse solute diffusion, further membrane development, concentration polarization, and the improvement of the draw solute design.

4.1. Concentration polarization mechanism in forward osmosis

When it comes to the osmotically driven and pressure-driven membrane processes, the concentration polarization is an inevitable and frequent phenomenon [11, 38–42]. As illustrated in **Figure 2**, in the osmotically driven membrane processes, the concentration polarization is produced by the overall concentration variance occurring between the draw solution and the feed solution through the asymmetric FO membrane. The internal concentration polarization (ICP) and external concentration polarization (ECP) can happen during the FO processes. In general, ICP happens within the membrane's porous support layer, and ECP happens at the surface of the membrane's dense active layer. The sections below further describe both ECP and ICP.
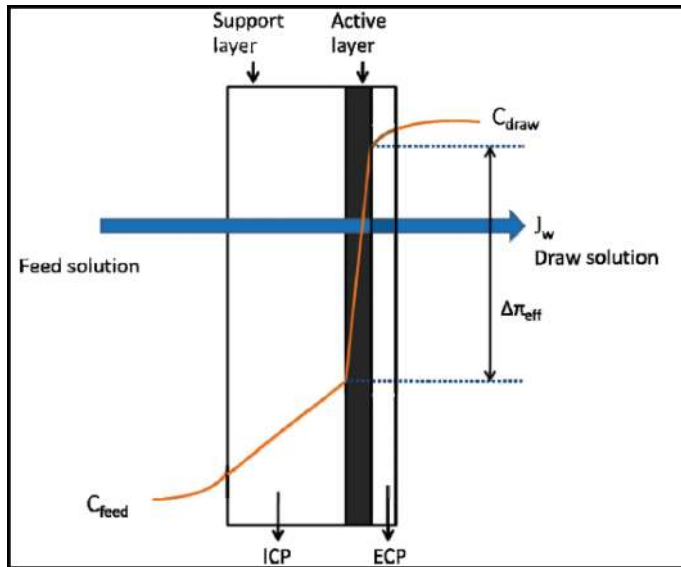
Figure 2. Internal concentration polarization (ICP) and external concentration polarization (ECP) through an asymmetric FO membrane [16].

4.1.1. External concentration polarization: modeling and mechanism

ECP in FO occurs at the surface of the membrane's active layer, similar to the other pressure-driven membrane processes. Their distinction is due to the fact that only concentrative ECP can occur in a pressure-driven membrane process, and both dilutive ECP and concentrative ECP can happen in an osmotically driven membrane process, conditional on the membrane's orientation with regard to the feed and the draw solutions. The dilutive ECP happens when the membrane's support layer is facing the feed solution, while the concentrative ECP occurs in instances where the membrane's support layer is facing the draw solution. ECP lowers the overall driving force due to the higher osmotic pressure at the membrane's active layer interface located on the membrane's feed side, or the lowered osmotic pressure at the membrane's active layer surface located on the draw solution side. The unfavorable effects of ECP on the permeate flux can be alleviated by optimizing the water flux and raising the flow's velocity or turbulence [11]. With the application of the boundary-layer film theory McCutcheon and Elimelech have successfully modeled ECP in FO [38, 43]. The generalized equation for concentration polarization modulus in pressure-driven membrane processes may be expressed in Eq. (5), as follows.

$$\frac{C_m}{C_b} = exp\left(\frac{J_w}{k}\right) \tag{5}$$

where J_w is the water flux, k is the mass transfer coefficient value, and C_m and C_b are the concentrations of the feed solution at the membrane's surface and in the bulk, respectively. The mass transfer coefficient (k) is related to the Sherwood number (Sh) by:

$$k = \frac{ShD}{D_h} \tag{6}$$

where D is the solute diffusion coefficient value, and D_h is the hydraulic characteristic length. When the feed solution concentration is relatively low in FO, the concentrations in Eq. (5) can be substituted by the osmotic pressures. As a result, the concentrative ECP modulus can be expressed as follows:

$$\frac{\pi_{m-feed}}{\pi_{b-feed}} = exp\left(\frac{J_w}{k_{feed}}\right) \tag{7}$$

where k_{feed} is the mass transfer coefficient on the feed side, π_{m-feed} and π_{b-feed} are the osmotic pressures of the feed solution at the membrane's surface and in the bulk, respectively. Similarly, the dilutive ECP modulus in FO can be expressed as:

$$\frac{\pi_{m-draw}}{\pi_{b-draw}} = exp\left(-\frac{J_w}{k_{draw}}\right) \tag{8}$$

where k_{draw} is the mass transfer coefficient on the draw side, and π_{m-draw} and π_{b-draw} are the osmotic pressures of the draw solution at the membrane's surface and in the bulk, respectively. Eqs. (1) and (2) reflect the water transport in RO, FO, and pressure-retarded osmosis (PRO), as indicated in Eqs. (1) and (2), as shown in Section 3. Both π_{draw} and π_{feed} should be the effective osmotic pressures at the membrane's surfaces, specifically.

$$J_w = A\left(\pi_{m-draw} - \pi_{m-feed}\right) \tag{9}$$

By substituting Eqs. (7) and (8) into Eq. (9), Eq. (10) can be obtained as below:

$$J_w = A\left[\pi_{b-draw}\ exp\left(-\frac{J_w}{k_{draw}}\right) - \pi_{b-feed}\ exp\left(\frac{J_w}{k_{feed}}\right)\right] \tag{10}$$

Although the dilutive ECP and concentrative ECP have been examined in Eq. (10) [43], there are multiple key points that have to be noted in Eq. (10). First of all, the mass transfer coefficient values on the feed and draw solution sides are not the same because of the varying hydraulic conditions between the draw solution side and the feed side. Next, this model relies on multiple assumptions, including that the solute permeability's coefficient is zero (i.e., the reflection coefficient σ = 1 [44]) and that the draw and feed solution concentration values are reasonably low, since only in this case can it be accepted that the concentration is equal to the corresponding osmotic pressure values. Finally, it must be noted that this model is adequate only in instances with a dense symmetric film, instead of an asymmetric-type membrane. As a result, the uses for this model can be somewhat limited. It is necessary to examine the dynamic where an asymmetric FO membrane is applied in a manner that would replicate its real-world practical uses and where the ICP effects become more significant.

4.1.2. Internal concentration polarization: modeling and mechanism

ICP is a critical aspect of the osmotically driven membrane-type processes. Research indicates that the water flux decline in FO is primarily produced by ICP [38, 44–46]. The early research

projects that looked at FO suggested that ICP might lower the water flux by more than 80% [45, 47]. As indicated in **Figure 3**, there are two types of ICP, concentrative ICP and dilutive ICP, occurring within the membrane's support layer, and they depend on the membrane's orientation [48]. Once the draw solution is situated against the membrane's support layer, dilutive ICP can successfully happen within the membrane's support layer since the water permeates across the membrane, from the feed solution to the draw solution. In a different membrane orientation where the feed solution is opposite the membrane support layer, concentrative ICP happens when the solute properly accumulates within the membrane's support layer located on the feed side. The ICP process is happening in the support layer and, as a result, it cannot be weakened through a change in the hydrodynamic conditions, including higher turbulence or flow rate.

The effects of ICP on FO water flux have been modeled using an adaptation of the classical solution-diffusion theory [38, 43]. The dilutive ICP dominates the water flux (Jw) when the draw solution is placed against the membrane support layer (i.e., FO mode) and can be expressed [49] as follows:

$$J_w = \frac{1}{K} ln \frac{A\pi_{draw} + B}{A\pi_{feed} + B + J_w} \tag{11}$$

where B is the membrane's solute permeability coefficient, and K is the solute resistivity value, a measure of solute transport in the membrane's support layer. K is used to quantify the solute's capacity to diffuse into or out of the membrane's support layer, and it can reflect the degree of ICP available in the support layer. Lower K values indicate less ICP and cause greater pure water flux (J_w). K is defined earlier in Eq. (4). It should be noted that the structural parameter S, in Eq. (4), is an essential membrane quality since it governs ICP in the membrane's support by establishing membrane's tortuosity, porosity, and thickness values. As a
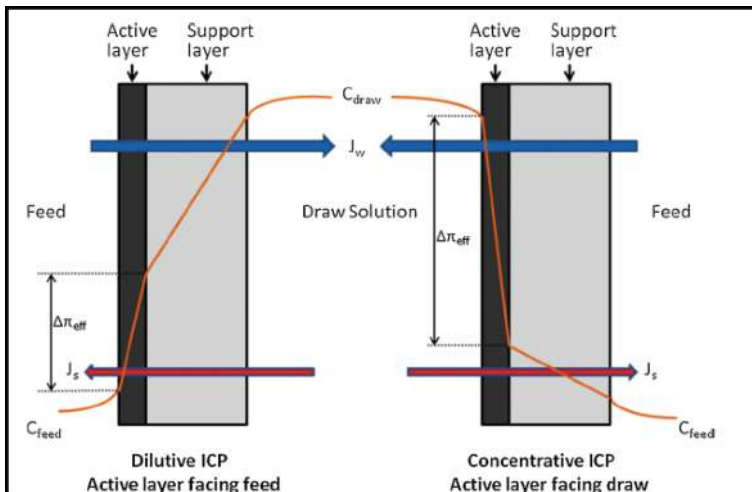

Figure 3. Dilutive ICP and concentrative ICP across an asymmetric FO membrane [48].

result, when it comes to the newly developed membranes, it is crucial to describe the membrane's structural parameter S. The value of S can be calculated based on Eqs. (4) and (11) and by fitting FO test results [50, 51]. On the other hand, in a specific membrane orientation, both ICP and ECP happen concurrently rather than occurring separately. Researchers McCutcheon and Elimelech have designed models that consider the characteristics and influences of both ECP and ICP. For the FO mode, the analytical model capturing the effects of both concentrative ECP and dilutive ICP on permeate flux may be conveyed [38, 43] by:

$$J_w = A \left[\pi_{draw} \; exp \left(-J_w K \right) - \pi_{feed} \; exp \left(\frac{J_w}{k} \right) \right] \tag{12}$$

As seen in Eq. (4), it appears that ICP in the membrane's support layer is formed based on the membrane properties, such as membrane's tortuosity, porosity, and thickness, as well as the diffusion solute properties, like the diffusion coefficient of the solute. A research project by Zhao and Zou has connected ICP to additional properties of the solution, like viscosity and diffusion solute size, by considering the idea of constrictivity [48]. The equation that corresponds to this dynamic is embodied in the following:

$$K = \frac{t\tau}{\delta \varepsilon_{eff} D} \tag{13}$$

In this case, a new parameter δ is expressed as the constrictivity factor, and ε_{eff} is the effective transport through porosity, as it can be lower than the overall membrane porosity if certain small pores are not available to the larger solute. In particular, the constrictivity parameter relies on the ratio of the pore diameter to the solute molecule diameter:

$$\lambda = \frac{molecule \; diameter}{pore \; diameter} < 1 \tag{14}$$

Tang et al. researched the cumulative effect of fouling and ICP on FO flux behavior. [50]. Tang et al. noted two critical phenomena during the experimental runs. The first phenomenon was that the water flux was comparatively stable and its decrease was minor during the FO mode, whereas during the PRO mode, the flux decrease was substantial and especially prominent when membrane fouling happened. The second phenomenon had to do with the fact that the effects of ICP on FO flux were more distinct at greater draw solution concentration values [48]. A number of new modeling techniques have been used to research the concentration polarization (CP) phenomenon, such as the computational fluid dynamics (CFD) [52], numerical simulation [53], and the finite element method (FEM) [54, 55]. A project spearheaded by Li et al. used FEM to interrogate the relationship between the membrane's porous structure and ICP [54]. The mathematical models that came out of this project can serve as a valuable toolkit for improving FO performance and optimizing the membrane's support construction [54].

4.2. Membrane fouling mechanism in forward osmosis

Like concentration polarization, membrane fouling is an unavoidable as well as essential phenomenon influencing all types of membrane processes [28, 56–63]. As a consequence,

smaller membrane fouling potential ensures that there is less cleaning, longer membrane life, and more water produced, which effectually decreases capital and operational costs. On the other hand, the membrane-type fouling happening in osmotically driven membrane processes is distinct from the types of fouling present in pressure-driven membrane processes, as a result of the low hydraulic pressure being used in the former case. Initially, Cath et al. researched membrane fouling in FO in relation to systems used in long-term space missions [64, 65]. Cath et al. suggested that FO could have the capacity to reduce membrane fouling, since there was no flux decrease due to fouling detected during the experimental runs [65]. During the last few years, FO has been applied in osmotic membrane bioreactors (OMBR) primarily for wastewater treatment because of its lower energy consumption and lower fouling needs [20, 22], both of which are two challenges for membrane bioreactors [59, 66]. In a research project by In addition, the OMBR system was used to treat activated sludge. The results report that neither irreversible nor reversible fouling was seen whenever the membrane's active layer was positioned in a way facing the activated sludge [22]. An experiment conducted by Achilli et al. relied on a submerged OMBR so as to treat domestic wastewater over the prolonged period of up to 28 days, indicating that the decrease of water flux was primarily due to membrane fouling [20]. On the other hand, the flux of the initial values could be recovered by roughly 90% through the process of osmotic backwashing. This experimental result suggests that membrane fouling in OMBR may in fact be reversible. Similarly, the data reflect that membrane fouling does exist in FO and is apparent during long-term operational runs. Mi and Elimelech interrogated the inorganic and organic fouling in FO [23, 27, 62]. Mi and Elimelech determined that, first of all, the intermolecular adhesion and organic fouling were connected and that foulant-foulant interactions had an important role in organic cleaning and fouling. Second, Mi and Elimelech found out that FO fouling was controlled by the coupled effects of chemical, for example, calcium binding, and hydrodynamic, for instance permeation drag and shear force, interactions. They likewise noted that membrane materials had a key role in organic fouling and cleaning, which was later verified with the help of atomic force microscope (AFM) measurements. Mi and Elimelech also found that both inorganic and organic types of fouling in FO were nearly fully reversible using water rinsing. This could be attributed to the less compact fouling layer created by the applied low hydraulic pressure, which suggests that chemical cleaning could be prevented. Moreover, researchers comparing membrane fouling in FO and RO suggested that it could be diverse from one case to another with respect to water cleaning efficiency and reversibility [23, 27, 28]. Although it was irreversible in RO, Lee et al. observed that membrane fouling in FO was almost entirely reversible [28]. Alternatively, Lee et al. linked the FO fouling to the accelerated cake-enhanced osmotic pressure (CEOP) created by the reverse solute (salt) diffusion process in the draw solution. **Figure 4** outlines the mechanics of this process [31]. Once the draw solution faces the membrane's support layer, using the reverse diffusion, the draw solute collects on the active layer's surface located on the feed side, lowering the net osmotic driving force and improving the concentration polarization layer. The draw solute featuring a less hydrated radius value (e.g., NaCl) is more easily capable of initiating CEOP, when compared to the ones with a greater hydrated radius values, like dextrose. In an experiment by Lay et al., it was noted that the reverse diffusion of the draw solute could worsen the CEOP effect as well as intensify FO fouling [67]. Alternatively, new research suggests that FO fouling could be substantially lowered if the cross flow velocity is increased [28].
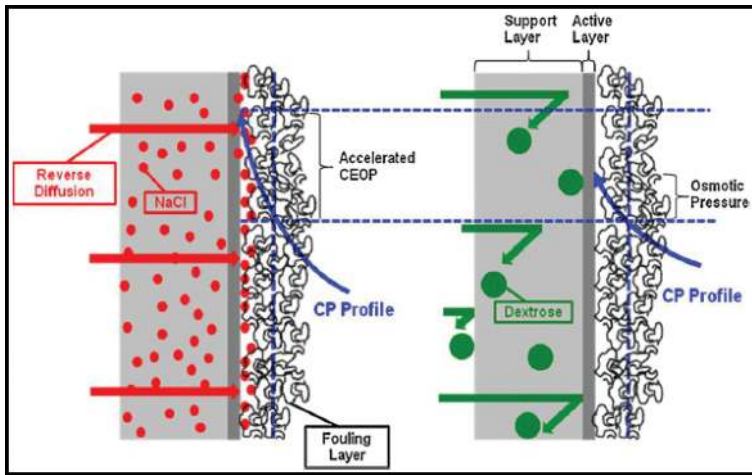
Figure 4. The effects of draw solute reverse diffusion on cake-enhanced osmotic pressure (CEOP) in FO for two different draw solutions: (a) NaCl and (b) dextrose [31].

In a recent experimental attempt, Tang's group used direct microscopic observation to study FO fouling and its mechanisms. They determined that the critical flux concept could also be relevant to osmotically drive the types of processes [63]. Direct microscopic observation has been implemented to research the production of fouling in pressure-driven membrane processes and currently remains one of the primary membrane fouling characterization methodologies [68–71]. Admittedly, direct microscopic observation is relevant only for the cases with large foulants in colloidal or biofouling fouling, such as microbes or particles. Thus, direct microscopic observation can likewise be implemented in the research of membrane fouling if visible fouling layers or large foulants are present in FO.

Usually defined as the level of permeate flux where membrane fouling becomes noticeable, critical flux has been widely used in pressure-driven membrane processes [72–75]. Critical flux can also be applied to osmotically driven membrane processes. A recent study by Zhao et al. confirms its presence in FO [76]. It is necessary to note that the critical flux identified by Zhao et al.'s FO research study, as well as by Tang and coworkers, was detected when the membrane's surface was partially covered with visible foulant [63, 76]. As a consequence, the critical flux in FO could have an implicit connection to the visible fouling layer. This particular connection must be investigated in greater depth. Research suggests that greater working temperatures can have various negative influences on FO cleaning and scaling in brackish water desalination processes, potentially because of the change of HCO_3^- into CO_3^{2-} at high temperature values [26]. The report indicates that, caused by the polymerization of dissolved silica, the silica scaling of FO membranes was the primary inorganic type of fouling in real-case seawater desalination examples [77]. The silica polymerization might likewise quicken the organic fouling, which is removed much easier using water rinsing if compared to the silica scaling [77].

Alternatively, membrane fouling could improve the FO membrane's solute rejection potential. It was also detected that organic foulants located on the membrane's surface, or its active layer,

could improve the negative charge property and surface hydrophilicity, and this in turn can raise the hydrophilic compound absorption capacity [78]. Changes like these can increase the critical rejection potential for many new contaminants, including trace organic compounds, as well as hydrophobic neutral compounds and hydrophilic ionic compounds [78]. Once the FO tests were run continuously for prolonged periods of time at the pilot scale, the rejection performance values improved even further as more substantial fouling happened [79]. In a project by Jin et al., it was determined that organic fouling can likewise have substantial consequences for the elimination of inorganic contaminants, like arsenic and boron [80]. In particular, their influences relied on the membrane's orientation. For instance, in the FO mode, where the membrane's support layer is facing the draw solution, the organic fouling on the membrane's active layer can improve the sieving influence and essentially increase the arsenic rejection in the feed. Alternatively, in the PRO mode, where the membrane's support layer is facing the feed, the organic fouling in the membrane's support structure can lower the boron rejection [80]. Membrane fouling and concentration polarization remain critical phenomena in FO processes since they have the capacity to heighten the additional membrane resistance and lower membrane permeability potential. Researchers must continue to further examine their functions and mechanisms if they want to improve the FO process and its performance capacity. Successful application of FO in real settings will remain problematic until a more comprehensive analysis becomes available.

4.3. Reverse solute diffusion

In membrane processes that are osmotically driven, the solute's reverse diffusion, from the draw solution and through the membrane toward the feed solution, is likewise almost certainly due to the concentration variances. Cath et al. (2009) state that the draw solute reverse diffusion has to be carefully studied as it could endanger the success of the process [11, 81]. Some research studies have linked draw solute reverse diffusion with the membrane fouling phenomenon. Lay et al. and Lee et al. have shown that the draw solute reverse diffusion can, on the one hand, improve the CEOP influence and, on the other hand, intensify FO fouling [28, 67]. Thus, multivalent ion solutions featuring smaller diffusion coefficient values are better for certain uses in which higher rejection potentials are required [11]. Alternatively, in other cases, multivalent ions, like Ca^{2+} and Mg^{2+}, could impede the foulants in the feed solution following reverse diffusion, a dynamic that can worsen the overall membrane fouling [82]. Furthermore, multivalent ions could likewise incite a more substantial ICP due to their smaller diffusion coefficients and bigger ion sizes [48]. Defined as the ratio of the reverse solute flux to the forward water flux, specific reverse solute flux has been added as another potential measure of membrane's selectivity [81, 82]. Specific reverse solute flux parameter offers a third dynamic for the proper FO performance evaluation, together with the salt rejection and the permeate flux parameters. A greater specific reverse solute flux suggests reduced membrane selectivity potential, as well as an inferior FO efficiency value. Although the specific reverse solute flux depends on the membrane's active layer selectivity, it is independent of the structure of the membrane support layer and the draw solution concentration values [51]. This key outcome grants another standard for the production of a new type of FO membrane, that is, greater selectivity of the membrane's active layer. Moreover, engaging a multivalent ion solution as the draw solution could reduce membrane fouling [28, 67] and lower the reverse solute

diffusion [81], but in this case, there is also a potential to have a higher ICP [48] and a greater risk of fouling [82]. To sum up, reverse solute diffusion remains one of the main challenges in osmotically driven membrane processes and as a result must be reduced during the production and design of draw solutes and FO membranes.

4.4. Membrane development

Based on the available membrane fabrication methodologies, newly produced and designed membranes can be organized into three categories: the thin film composite (TFC) membranes, the chemically modified membranes, and the phase inversion-formed cellulosic membranes. Reverse solute diffusion, membrane fouling, and ICP are three of the crucial concerns that exist with respect to the osmotically driven membrane processes, since they effectually direct the FO performance. As a result, when considering innovative FO membrane development, it is essential to characterize its salt rejection, antifouling, and anti-ICP characteristics. When compared with other types of processes, FO could be viewed as more competitive when treating challenging waters with higher fouling potential or solid content, since ICP and fouling are frequently much more serious. When it comes to FO, the water flux is affected by the water permeability, while the reverse solute flux is shaped by the membrane solute permeability. In this instance, there is a type of trade-off between salt rejection and water permeability [83]. Higher water permeability values are desirable, as well as lower salt rejection potential. In most cases, FO membrane featuring higher water permeability potential likewise offers higher salt flux, and the reverse relationship holds true as well. As a consequence, defined as the ratio of the reverse solute flux to the forward water flux, specific reverse solute flux can be a superior parameter to evaluate when considering the FO performance [81]. In fact, it might be better to assess the FO performance with the aid of the osmotic water flux and specific reverse solute flux when membrane fouling and ICP are present. Thus, the characterization and design of new FO membrane in the forthcoming future must reflect on the antifouling and the anti-ICP properties, as well as salt rejection (solute permeability), structural parameters, and water permeability.

4.4.1. Phase inversion-formed cellulosic membranes

Asymmetric cellulosic osmotically driven membranes developed through phase inversion have been created specifically for osmotic drug delivery before they were used for water treatment purposes [33, 84, 85]. Most of these membranes were created using conventional phase inversion and with the help of cellulose acetate as the dip-coating polymer. A research breakthrough in Loeb and Sourirajan's method occurred when they prepared RO membranes through phase inversion based on cellulose acetate polymer. Cellulose acetate offers a variety of desirable properties, such as a comparatively high hydrophilicity favoring lower fouling propensity and greater water flux, wide availability, improved mechanical strength, as well as enhanced resistance to degradation by chlorine and other types of oxidants [86, 87]. This particular form of cellulosic membrane is implemented in energy generation, such as osmotic power, and through a PRO process [88]. Recently, Chung's research group has produced a number of cellulose ester-based membranes specifically for FO applications and containing flat sheet modules and hollow fiber [89–91]. In this case, the methods for creating these cellulose derivative membranes are relatively similar, in the form of phase inversion that is followed by hot water annealing at 60–95°C. Chung's research group determined that the resulting

membrane could have two selective skin layers that are capable of lowering ICP in the membrane support layer [87, 90]. A more recent study modeled this type of double-skinned FO membrane [92]. Chung's research group likewise noted that the relationship between the casting substrate and the polymer had an important role during phase inversion for the development of the membrane's structure [87, 91]. Furthermore, Sairam et al. implemented this phase inversion approach in order to create flat sheet FO membranes using cellulose acetate [93]. Specifically, they applied maleic acid, zinc chloride, and lactic acid as pore-forming agents, while casting the membrane onto nylon fabric at a range of annealing temperature values. Sairam et al. noted that the membrane developed with zinc chloride as the pore-forming agent allowed for a reasonably effective FO performance. On the other hand, the disadvantages of cellulose acetate have to be examined before it is used in FO membranes. While cellulose acetate membranes are more resistant to chloride degradation and more hydrophilic, if compared to the TFC polyamide RO membranes, they have lower resistance potential to biological attachments and hydrolysis [86, 94, 95]. To reduce the hydrolysis of cellulose acetate membranes, it is crucial to modify the pH of the feed and draw solutions within the ranges of 4–6 and to sustain the working temperature that does not rise above 35°C [86, 94].

4.4.2. Thin film composite membranes

It has been noted that there is a key trade-off dynamic occurring between salt rejection and water permeability potential. For instance, the raising of the trimesoyl chloride (TMC) concentration or the reduction of the m-phenylenediamine (MPD) concentration caused greater membrane permeability potential but lower salt rejection values [83]. Research likewise indicates that greater hydrophilicity of the support layer may prefer water diffusion across the FO membrane [96, 97]. Wang et al. prepared polyethersulfone (PES)/sulfonated polysulfone (PSF)-alloyed–type membranes as the substrates of interfacial polymerization and produced high-performance FO membranes. On the other hand, Yu et al. developed a nonporous polyethersulfone (PES) FO-type membrane with the aid of phase inversion, however, without using interfacial polymerization [98]. In this case, the polyester nonwoven fabrics were implemented for backing support. This membrane creation approach was comparable to the one used by Elimelech's group, with the exception of the additional interfacial polymerization phase. According to the report, the membrane produced by this method featured an active layer formed on top of the support layer, high water flux value, and low reverse solute flux potential [98].

Song et al. reported the creation of a nanofiber TFC FO-type membrane using electrospinning, which was followed by interfacial polymerization (ES-IP) [99]. Song et al. noted that the nanocomposite FO membrane allowed for an improved FO performance mostly because of high porosity and low tortuosity that significantly decreased the structural parameters of the membrane. If compared to the TFC FO membrane made using phase inversion followed by interfacial polymerization (PI-IP), the electrospinning-formed nanofiber support layer offers a porous structure resembling a scaffold with interlocked pores between individual nanofibers [99]. Due to this structure, the water flux value of the ES-IP–formed FO membrane was found to be three times as high as the water flux potential of the PI-IP–formed membrane. In this instance, the performance of the FO membrane was enhanced with respect to osmotic water flux, while salt rejection was obtained as well as confirmed by Bui et al. research group [100]. The majority of the approaches used for preparing TFC FO membranes and asymmetric

cellulose acetate FO membranes are indistinguishable from the original traditional RO membrane methods, like phase inversion followed by interfacial polymerization, or phase inversion and annealing.

When it comes to a TFC FO membrane, the membrane support layer made using phase inversion governs the ICP, water flux in FO, and the membrane's active layer controls reverse solute flux potential and salt rejection values. A high salt rejection can be obtained when the TFC membranes are developed with the help of interfacial polymerization. In fact, FO's performance is shaped by the membrane's support layer. Next-generation FO membrane production must pay attention to the membrane's support layer and its role. All in all, an effective FO membrane has to provide a design that appears sufficiently porous and offers improved hydrophilic support combined with lower tortuosity capable of decreasing ICP, as well as a selective active layer that can lower reverse solute diffusion and augment salt rejection potential.

4.4.3. Chemically modified membranes

Over the course of the last several years, chemical modification methodologies have been implemented during the development of innovative FO membranes. Arena et al. research group (2011) applied polydopamine (PDA) as a new bioinspired hydrophilic polymer for the modification of the support layers in commercial TFC RO membranes catering to engineered osmosis applications [101]. This modified membrane showed improved water flux and lower ICP during the conducted FO tests. Furthermore, Setiawan et al. created a hollow-type fiber FO membrane featuring a positively charged NF-like selective layer using a polyelectrolyte posttreatment of a polyamide-imide (PAI) microporous substrate with polyethylenimine (PEI) [102]. Setiawan et al. indicated that the final FO membrane produced could be applied in heavy metal removal processes due to its unique positively charged characteristic. This research group likewise designed a flat sheet–type membrane offering a positively charged NF-like selective layer on top of a woven fabric–embedded substrate and implementing a similar methodology. The reported results suggest that the overall thickness of the substrate was reduced to 55 µm when the PAI microporous substrate was successfully embedded within a woven fabric. Moreover, Tang and coworkers relied on a creative layer-by-layer assembly approach in order to produce FO membranes with desirable properties [103, 104]. In Tang's research studies, polyacrylonitrile (PAN) substrate was prepared with the aid of phase inversion and then posttreated by sodium hydroxide so as to improve surface negative charge density and hydrophilicity potential. Poly(sodium 4-styrene-sulfonate) (PSS) and poly(allylamine hydrochloride) (PAH) were implemented as the polyanion and polycation, respectively. Arguably, the majority of the present approaches to the FO membrane preparation are established methods that have been applied during the last few decades for the creation of pressure-driven–type membranes, such as RO and NF. The production and design of innovative high-performance FO membranes are still in their early stages. As a result, the process of relying on the older methodologies for RO or NF membrane preparation is a sensible and practical direction. Forthcoming research may expand the recently developed techniques for the production of high-performance FO membranes, including layer-by-layer assembly [103, 105–112], UV-photographing [113–116], and polyelectrolyte dip-coating [117, 118]. In addition, membranes featuring polyelectrolyte multilayers, charged properties, or double selective layers can provide exciting avenues for specific real-life FO applications.

4.5. Draw solute/solution developments

Despite preexisting setbacks, key innovative approaches for draw solute selection have been suggested [119, 120]. Specifically, there are three conditions for successful selection of a suitable draw solution in FO applications. To begin, the draw solution must offer a reasonably high osmotic pressure values [11]. Next, the diluted draw solution needs to be economically and effortlessly reconcentrated and recovered [11, 121]. Finally, the draw solute has to showcase lowered ICP during the FO processes. A research study by Zhao and Zou indicates that ICP in FO is seriously influenced by draw solution viscosity, draw solution's ion/molecule size of the solute, and solute diffusion coefficient values [48]. Increased diffusion coefficients, coupled with lowered ion/molecule sizes and smaller solution viscosities, will minimize ICP and allow for resulting in improved permeate fluxes [48]. Moreover, additional parameters like low reverse solute permeability [11], zero toxicity, low cost, absence of membrane damage, inertness and stability at or near natural pH, and good biofouling-resistance should be considered when the draw solute/solution is selected.

During the last few decades, numerous draw solutes/solutions have been examined during osmotically driven–type processes. The primary benefit of implementing volatile gases as draw solutes in FO is that the final thermolytic draw solution may be separated or recovered with the help of heating/or distillation. In a separate project, sugars were likewise tested as draw solutes since there is no necessity to separate the diluted nutrient solutions further, and the diluted solutions may be reconcentrated in decreased pressures with loose RO membranes. After the 2000s, Elimelech and coworkers suggested a new draw solution for the purposes of FO desalination, that is, a water-soluble mixture of NH_3 and CO_2 including ammonium bicarbonate (NH_4HCO_3) [9, 10, 24]. The proposed draw solution can offer improved water fluxes as a result of the higher driving forces created by the greater solubilities of the solutes. These types of draw solutes may be effortlessly recovered or recycled using moderate heating ($\sim 60°C$) [9]. As a result, this innovative draw solution could find a potential application in large-scale desalination, even though the removal the ammonia (NH_3) smell from the produced water could be a concern. Furthermore, various other chemicals have been assessed for the role of the draw solutes [119].

For instance, synthetic materials, like organic compounds [121] and magnetic and/or hydrophilic nanoparticles [122–124], have been proposed for the application as the draw solutes. In the case of the laboratory-designed magnetic nanoparticles, data suggest that the particle size and surface hydrophilicity of the particles had critical roles for the FO separation performance [123]. It was also noted that particle agglomeration happened during draw solute recycling process using magnetic separators [124]. Such an accumulation of magnetic nanoparticles may be reduced with the aid of ultrasonication. When using this method, the particles' magnetic characteristics and the recovery efficacy were threatened by ultrasonication as well. In order to transcend the issue of accumulation during draw solute recycling, the thermal-responsive properties were integrated into the magnetic nanoparticles using the one-step thermal decomposition [125]. UF could likewise be used to recover diluted draw solutes featuring big particle or molecule sizes. Wang's research group has produced a stimuli-responsive polymer hydrogel as another draw solute for FO desalination [126]. Polymer hydrogels such as these have the capacity to pull water from the saline feed during swelling and after that release the water while deflating, the latter being caused by heating and hydraulic pressure. So as to enhance the

capacity of swelling ratios and drawing water, a type of light-absorbing carbon particles was introduced into the polymer hydrogels, and as a consequence an improved performance was obtained [127]. A new draw solute separation system simulating the "destabilization" phenomenon was suggested for the process of generating drinking water [128]. In this particular system, $Al_2(SO_4)_3$ was chosen as the draw solute, while the diluted $Al_2(SO_4)_3$ solution pH was attuned using CaO, finally resulting in the creation of a white gel-like mixture made out of positively charged $Al(OH)_3$ and $CaSO_4$. In the following step, negatively charged superparamagnetic nanoparticles were added so as to enable the sedimentation. To accelerate the sedimentation process and enhance separation efficiency, an external magnetic field was introduced. Such an innovative draw solution separation technique can make FO a more economical and eco-sustainable process for efficient drinking water production [128].

The selection criteria for the draw solutes and solutions need to be addressed for the process to be effective and sustainable. An effective draw solute option for FO must offer easy and economical recovery, lower tendency to cause ICP, zero toxicity, reasonable costs, higher solubility, and greater osmotic pressure. The diffusion coefficient, viscosity of the draw solution, and the solute particle size need to be examined as they are directly connected to ICP [48] effectually dominating the water flux in FO [38, 46].

4.6. Forward osmosis fouling control

In this chapter, the focus was on discussing and reviewing the primary five issues that exist in FO. Certainly, these challenges do not exist in isolation but are rather interconnected. To sum up some of these issues, the membrane's support layer needs to be as porous as possible for the lower ICP, and the membrane's active layer needs to be more selective for a lower reverse solute diffusion potential. The smaller reverse solute diffusion can then decrease the membrane fouling. When it comes to the draw solute, small ion or molecule sizes could minimize ICP [48]; however, they can likewise increase membrane fouling and reverse solute diffusion. All of these correlations and criteria make the creation of advantageous draw solutes much more problematic. In most cases, higher reverse solute diffusion may lead to substantial membrane fouling, and this correlation holds the other way as well [28, 67]. In addition, ICP and membrane fouling could lead to multiple adverse properties for water flux in FO [50]. Furthermore, reverse solute diffusion, membrane fouling, and ICP are at their core determined by draw solute properties and membrane qualifications.

5. Conclusion

The membrane processes based on osmosis are new technological directions that have exhibited a lot of promise for a range of applications, and especially water purification, food processing, desalination, wastewater treatment, power generation, and pharmaceutical product dehydration. While FO is not likely to fully replace RO as the primary desalination technology in the foreseeable future, it remains an appealing alternative as an effective desalination approach offering many benefits over pressure-driven–type membrane processes. In order to transfer FO from the laboratory stages of research into hands-on industrial applications, a set of advances in terms of FO membrane and draw solute development needs to happen. In fact, the membranes

need to offer critical properties of minimizing ICP, higher mechanical strength, stability, improved water permeability, and better selectivity. To sum up, this chapter examined five essential challenges for FO in the form of membrane fouling, reverse solute diffusion, further membrane development, concentration polarization, and enhanced draw solute design. The innovative draw solutes must be capable of producing higher osmotic pressure, remain easily and economically regenerated/or recycled, and provide minimal ICP. Draw solutes must also offer compatibility with the FO membranes and zero toxicity. A successful draw solute has a vital role in the popularization and efficacy of FO applications. The next level of draw solute development will allow for a much wider use of FO in a range of industrial-scale applications and fields.

Nomenclature

A	water permeability constant of the membrane
B	solute permeability coefficient of the membrane
C_m	concentrations of the feed solution at the membrane surface
C_b	concentrations of the feed solution at the bulk
D	diffusion coefficient of the solute
D_h	hydraulic diameter
J_w	water flux
k	mass transfer coefficient
k_{feed}	mass transfer coefficient on the feed side
k_{draw}	mass transfer coefficient on the draw side
t	thickness of the membrane
τ	tortuosity
σ	reflection coefficient
ε	membrane porosity
S	membrane structural parameter
Sh	Sherwood number
ΔP	applied hydraulic pressure difference
$\Delta\pi$	osmotic pressure difference across the membrane
$\Delta\pi_{Feed}$	bulk osmotic pressure of the feed solution
π_{m-draw}	osmotic pressures of the draw solution at the membrane surface
π_{b-draw}	osmotic pressures of the draw solution in the bulk
$\Delta\pi_{Draw}$	bulk osmotic pressure of the draw solution

Abbreviations

AFM   atomic force microscope

CEOP   cake-enhanced osmotic pressure

CFD   computational fluid dynamics

ECP   external concentration polarization

ES-IP   electrospinning followed by interfacial polymerization

FEM   finite element method

FO   forward osmosis

ICP   internal concentration polarization

MPD   m-phenylenediamine

NF   nanofiltration

OMBR   osmotic membrane bioreactor

PAH   poly(allylamine hydrochloride)

PAI   polyamide-imide

PAN   polyacrylonitrile

PDA   polydopamine

PEI   polyethylenimine

PES   polyethersulfone

PRO   pressure-retarded osmosis

PSF   polysulfone

PSS   poly(sodium 4-styrene-sulfonate)

RO   reverse osmosis

TFC   thin film composite

TMC   trimesoyl chloride

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References

[1] Shannon MA, Bohn PW, Elimelech M, Georgiadis JG, Marinas BJ, Mayes AM. Science and technology for water purification in the coming decades. Nature. 2008;**452**: 301-310

[2] E.I.A. International. World Energy Outlook 2010. The Energy Information Administration. Washington DC; 2010

[3] Elimelech M, Phillip WA. The future of seawater desalination: Energy, technology, and the environment. Science. 2011;**333**:712-717

[4] Montgomery MA, Elimelech M. Water and sanitation in developing countries: Including health in the equation. Environmental Science & Technology. 2007;**41**:17-24

[5] Elimelech M. The global challenge for adequate and safe water. Journal of Water Supply: Research and Technology—AQUA. 2006;**55**:3-10

[6] Gleick PH. Water resources. In: Schneider SH, editor. Encyclopedia of Climate and Weather. New York: Oxford University Press; 1996

[7] King CW, Webber ME. Water intensity of transportation. Environmental Science & Technology. 2008;**42**:7866-7872

[8] Geise GM, Lee H-S, Miller DJ, Freeman BD, McGrath JE, Paul DR. Water purification by membranes: The role of polymer science. Journal of Polymer Science Part B: Polymer Physics. 2010;**48**:1685-1718

[9] McCutcheon JR, McGinnis RL, Elimelech M. A novel ammonia–carbon dioxide forward (direct) osmosis desalination process. Desalination. 2005;**174**:1-11

[10] McCutcheon JR, McGinnis RL, Elimelech M. Desalination by ammonia–carbon dioxide forward osmosis: Influence of draw and feed solution concentrations on process performance. Journal of Membrane Science. 2006;**278**:114-123

[11] Cath TY, Childress AE, Elimelech M. Forward osmosis: Principles, applications, and recent developments. Journal of Membrane Science. 2006;**281**:70-87

[12] Petrotos KB, Quantick P, Petropakis H. A study of the direct osmotic concentration of tomato juice in tubular membrane-module configuration. I. The effect of certain basic process parameters on the process performance. Journal of Membrane Science. 1998;**150**: 99-110

[13] Petrotos KB, Quantick PC, Petropakis H. Direct osmotic concentration of tomato juice in tubular membrane-module configuration. II. The effect of using clarified tomato juice on the process performance. Journal of Membrane Science. 1999;**160**:171-177

[14] Petrotos KB, Lazarides HN. Osmotic concentration of liquid foods. Journal of Food Engineering. 2001;**49**:201-206

[15] Garcia-Castello EM, McCutcheon JR, Elimelech M. Performance evaluation of sucrose concentration using forward osmosis. Journal of Membrane Science. 2009;**338**:61-66

[16] Lee KL, Baker RW, Lonsdale HK. Membranes for power generation by pressure-retarded osmosis. Journal of Membrane Science. 1981;**8**:141-171

[17] Seppälä A, Lampinen MJ. Thermodynamic optimizing of pressure-retarded osmosis power generation systems. Journal of Membrane Science. 1999;**161**:115-138

[18] McGinnis RL, Elimelech M. Global challenges in energy and water supply: The promise of engineered osmosis. Environmental Science & Technology. 2008;**42**:8625-8629

[19] Achilli A, Cath TY, Childress AE. Power generation with pressure retarded osmosis: An experimental and theoretical investigation. Journal of Membrane Science. 2009;**343**:42-52

[20] Achilli A, Cath TY, Marchand EA, Childress AE. The forward osmosis membrane bioreactor: A low fouling alternative to MBR processes. Desalination. 2009;**239**:10-21

[21] Holloway RW, Childress AE, Dennett KE, Cath TY. Forward osmosis for concentration of anaerobic digester centrate. Water Research. 2007;**41**:4005-4014

[22] Liu Y, Mi B. Combined fouling of forward osmosis membranes: Synergistic foulant interaction and direct observation of fouling layer formation. Journal of Membrane Science. 2012;**407-408**:136-144

[23] Mi B, Elimelech M. Organic fouling of forward osmosis membranes: Fouling reversibility and cleaning without chemical reagents. Journal of Membrane Science. 2010;**348**:337-345

[24] McGinnis RL, Elimelech M. Energy requirements of ammonia–carbon dioxide forward osmosis desalination. Desalination. 2007;**207**:370-382

[25] Martinetti CR, Childress AE, Cath TY. High recovery of concentrated RO brines using forward osmosis and membrane distillation. Journal of Membrane Science. 2009;**331**:31-39

[26] Zhao S, Zou L. Effects of working temperature on separation performance, membrane scaling and cleaning in forward osmosis desalination. Desalination. 2011;**278**:157-164

[27] Mi B, Elimelech M. Gypsum scaling and cleaning in forward osmosis: Measurements and mechanisms. Environmental Science & Technology. 2010;**44**:2022-2028

[28] Lee S, Boo C, Elimelech M, Hong S. Comparison of fouling behavior in forward osmosis (FO) and reverse osmosis (RO). Journal of Membrane Science. 2010;**365**:34-39

[29] Cartinella JL, Cath TY, Flynn MT, Miller GC, Hunter KW, Childress AE. Removal of natural steroid hormones from wastewater using membrane contactor processes. Environmental Science & Technology. 2006;**40**:7381-7386

[30] Cath TY, Hancock NT, Lundin CD, Hoppe-Jones C, Drewes JE. A multi-barrier osmotic dilution process for simultaneous desalination and purification of impaired water. Journal of Membrane Science. 2010;**362**:417-426

[31] Yang Q, Wang KY, Chung T-S. A novel dual-layer forward osmosis membrane for protein enrichment and concentration. Separation and Purification Technology. 2009;**69**:269-274

[32] Jiao B, Cassano A, Drioli E. Recent advances on membrane processes for the concentration of fruit juices: A review. Journal of Food Engineering. 2004;**63**:303-324

[33] Wang C-Y, Ho H-O, Lin L-H, Lin Y-K, Sheu M-T. Asymmetric membrane capsules for delivery of poorly water-soluble drugs by osmotic effects. International Journal of Pharmaceutics. 2005;**297**:89-97

[34] Shokri J, Ahmadi P, Rashidi P, Shahsavari M, Rajabi-Siahboomi A, Nokhodchi A. Swellable elementary osmotic pump (SEOP): An effective device for delivery of poorly water-soluble drugs. European Journal of Pharmaceutics and Biopharmaceutics. 2008;**68**:289-297

[35] Bamaga OA, Yokochi A, Zabara B, Babaqi AS, Hybrid FO. RO desalination system: Preliminary assessment of osmotic energy recovery and designs of new FO membrane module configurations. Desalination. 2011;**268**:163-169

[36] Tang W, Ng HY. Concentration of brine by forward osmosis: Performance and influence of membrane structure. Desalination. 2008;**224**:143-153

[37] Mehta GD. Further results on the performance of present-day osmotic membranes in various osmotic regions. Journal of Membrane Science. 1982;**10**:3-19

[38] McCutcheon JR, Elimelech M. Influence of concentrative and dilutive internal concentration polarization on flux behavior in forward osmosis. Journal of Membrane Science. 2006;**284**:237-247

[39] Zydney AL. Stagnant film model for concentration polarization in membrane systems. Journal of Membrane Science. 1997;**130**:275-281

[40] Sablani SS, Goosen MFA, Al-Belushi R, Wilf M. Concentration polarization in ultrafiltration and reverse osmosis: A critical review. Desalination. 2001;**141**:269-289

[41] Thorsen T. Concentration polarisation by natural organic matter (NOM) in NF and UF. Journal of Membrane Science. 2004;**233**:79-91

[42] Kim S, Hoek EMV. Modeling concentration polarization in reverse osmosis processes. Desalination. 2005;**186**:111-128

[43] McCutcheon JR, Elimelech M. Modeling water flux in forward osmosis: Implications for improved membrane design. AICHE Journal. 2007;**53**:1736-1744

[44] Su J, Chung T-S. Sublayer structure and reflection coefficient and their effects on concentration polarization and membrane performance in FO processes. Journal of Membrane Science. 2011;**376**:214-224

[45] Mehta GD, Loeb S. Performance of permasep B-9 and B-10 membranes in various osmotic regions and at high osmotic pressures. Journal of Membrane Science. 1978;**4**:335-349

[46] Gray GT, McCutcheon JR, Elimelech M. Internal concentration polarization in forward osmosis: role of membrane orientation. Desalination. 2006;**197**:1-8

[47] Mehta GD, Loeb S. Internal polarization in the porous substructure of a semipermeable membrane under pressure-retarded osmosis. Journal of Membrane Science. 1978;**4**:261-265

[48] Zhao S, Zou L. Relating solution physicochemical properties to internal concentration polarization in forward osmosis. Journal of Membrane Science. 2011;**379**:459-467

[49] Loeb S, Titelman L, Korngold E, Freiman J. Effect of porous support fabric on osmosis through a Loeb–Sourirajan type asymmetric membrane. Journal of Membrane Science. 1997;**129**:243-249

[50] Tang CY, She Q, Lay WCL, Wang R, Fane AG. Coupled effects of internal concentration polarization and fouling on flux behavior of forward osmosis membranes during humic acid filtration. Journal of Membrane Science. 2010;**354**:123-133

[51] Phillip WA, Yong JS, Elimelech M. Reverse draw solute permeation in forward osmosis: Modeling and experiments. Environmental Science & Technology. 2010;**44**: 5170-5176

[52] Gruber MF, Johnson CJ, Tang CY, Jensen MH, Yde L, Hélix-Nielsen C. Computational fluid dynamics simulations of flow and concentration polarization in forward osmosis membrane systems. Journal of Membrane Science. 2011;**379**:488-495

[53] Jung DH, Lee J, Kim DY, Lee YG, Park M, Lee S, Yang DR, Kim JH. Simulation of forward osmosis membrane process: Effect of membrane orientation and flow direction of feed and draw solutions. Desalination. 2011;**277**:83-91

[54] Li W, Gao Y, Tang CY. Network modeling for studying the effect of support structure on internal concentration polarization during forward osmosis: Model development and theoretical analysis with FEM. Journal of Membrane Science. 2011;**379**:307-321

[55] Sagiv A, Semiat R. Finite element analysis of forward osmosis process using NaCl solutions. Journal of Membrane Science. 2011;**379**:86-96

[56] Jarusutthirak C, Amy G, Croué J-P. Fouling characteristics of wastewater effluent organic matter (EfOM) isolates on NF and UF membranes. Desalination. 2002;**145**:247-255

[57] Seidel A, Elimelech M. Coupling between chemical and physical interactions in natural organic matter (NOM) fouling of nanofiltration membranes: Implications for fouling control. Journal of Membrane Science. 2002;**203**:245-255

[58] Hoek EMV, Elimelech M. Cake-enhanced concentration polarization: A new fouling mechanism for salt-rejecting membranes. Environmental Science & Technology. 2003; **37**:5581-5588

[59] Le-Clech P, Chen V, Fane TAG. Fouling in membrane bioreactors used in wastewater treatment. Journal of Membrane Science. 2006;**284**:17-53

[60] Ang WS, Elimelech M. Protein (BSA) fouling of reverse osmosis membranes: Implications for wastewater reclamation. Journal of Membrane Science. 2007;**296**:83-92

[61] Tang CY, Chong TH, Fane AG. Colloidal interactions and fouling of NF and RO membranes: A review. Advances in Colloid and Interface Science. 2011;**164**:126-143

[62] Mi B, Elimelech M. Chemical and physical aspects of organic fouling of forward osmosis membranes. Journal of Membrane Science. 2008;**320**:292-302

[63] Wang Y, Wicaksana F, Tang CY, Fane AG. Direct microscopic observation of forward osmosis membrane fouling. Environmental Science & Technology. 2010;**44**:7102-7109

[64] Cath TY, Gormly S, Beaudry EG, Flynn MT, Adams VD, Childress AE. Membrane contactor processes for wastewater reclamation in space: Part I. Direct osmotic concentration as pretreatment for reverse osmosis. Journal of Membrane Science. 2005;**257**:85-98

[65] Cath TY, Adams D, Childress AE. Membrane contactor processes for wastewater reclamation in space: II. Combined direct osmosis, osmotic distillation, and membrane distillation for treatment of metabolic wastewater. Journal of Membrane Science. 2005;**257**:111-119

[66] Meng F, Chae S-R, Drews A, Kraume M, Shin H-S, Yang F. Recent advances in membrane bioreactors (MBRs): Membrane fouling and membrane material. Water Research. 2009;**43**:1489-1512

[67] Lay WCL, Chong TH, Tang C, Fane AG, Zhang J, Liu Y. Fouling propensity of forward osmosis: Investigation of the slower flux decline phenomenon. Water Science and Technology. 2010;**61**:927-936

[68] Kang S-T, Subramani A, Hoek EMV, Deshusses MA, Matsumoto MR. Direct observation of biofouling in cross-flow microfiltration: Mechanisms of deposition and release. Journal of Membrane Science. 2004;**244**:151-165

[69] Wang S, Guillen G, Hoek EMV. Direct observation of microbial adhesion to membranes. Environmental Science & Technology. 2005;**39**:6461-6469

[70] Li H, Fane AG, Coster HGL, Vigneswaran S. Direct observation of particle deposition on the membrane surface during crossflow microfiltration. Journal of Membrane Science. 1998;**149**:83-97

[71] Li H, Fane AG, Coster HGL, Vigneswaran S. Observation of deposition and removal behaviour of submicron bacteria on the membrane surface during crossflow microfiltration. Journal of Membrane Science. 2003;**217**:29-41

[72] Bacchin P, Aimar P, Field RW. Critical and sustainable fluxes: Theory, experiments and applications. Journal of Membrane Science. 2006;**281**:42-69

[73] Zhang YP, Fane AG, Law AWK. Critical flux and particle deposition of bidisperse suspensions during crossflow microfiltration. Journal of Membrane Science. 2006;**282**:189-197

[74] Zhang YP, Fane AG, Law AWK. Critical flux and particle deposition of fractal flocs during crossflow microfiltration. Journal of Membrane Science. 2010;**353**:28-35

[75] Field RW, Wu D, Howell JA, Gupta BB. Critical flux concept for microfiltration fouling. Journal of Membrane Science. 1995;**100**:259-272

[76] Zhao S, Zou L, Mulcahy D. Effects of membrane orientation on process performance in forward osmosis applications. Journal of Membrane Science. 2011;**382**:308-315

[77] Li Z-Y, Yangali-Quintanilla V, Valladares-Linares R, Li Q, Zhan T, Amy G. Flux patterns and membrane fouling propensity during desalination of seawater by forward osmosis. Water Research. 2012;**46**:195-204

[78] Valladares Linares R, Yangali-Quintanilla V, Li Z, Amy G. Rejection of micropollutants by clean and fouled forward osmosis membrane. Water Research. 2014;**45**:6737-6744

[79] Hancock NT, Xu P, Heil DM, Bellona C, Cath TY. Comprehensive benchand pilot-scale investigation of trace organic compounds rejection by forward osmosis. Environmental Science & Technology. 2011;**45**:8483-8490

[80] Jin X, She Q, Ang X, Tang CY. Removal of boron and arsenic by forward osmosis membrane: Influence of membrane orientation and organic fouling. Journal of Membrane Science. 2012;**389**:182-187

[81] Hancock NT, Cath TY. Solute coupled diffusion in osmotically driven membrane processes. Environmental Science & Technology. 2009;**43**:6769-6775

[82] Zou S, Gu Y, Xiao D, Tang CY. The role of physical and chemical parameters on forward osmosis membrane fouling during algae separation. Journal of Membrane Science. 2011;**366**:356-362

[83] Wei J, Liu X, Qiu C, Wang R, Tang CY. Influence of monomer concentrations on the performance of polyamide-based thin film composite forward osmosis membranes. Journal of Membrane Science. 2011;**381**:110-117

[84] Lin Y-K, Ho H-O. Investigations on the drug releasing mechanism from an asymmetric membrane-coated capsule with an in situ formed delivery orifice. Journal of Controlled Release. 2003;**89**:57-69

[85] Herbig SM, Cardinal JR, Korsmeyer RW, Smith KL. Asymmetric-membrane tablet coatings for osmotic drug delivery. Journal of Controlled Release. 1995;**35**:127-136

[86] Baker RW. Membrane Technology and Applications. 2nd ed. Etobicoke, Canada: John Wiley & Sons, Ltd.; 2004

[87] Zhang S, Wang KY, Chung T-S, Chen H, Jean YC, Amy G. Well-constructed cellulose acetate membranes for forward osmosis: Minimized internal concentration polarization with an ultra-thin selective layer. Journal of Membrane Science. 2010;**360**:522-535

[88] Gerstandt K, Peinemann KV, Skilhagen SE, Thorsen T, Holt T. Membrane processes in energy supply for an osmotic power plant. Desalination. 2008;**224**:64-70

[89] Su J, Yang Q, Teo JF, Chung T-S. Cellulose acetate nanofiltration hollow fiber membranes for forward osmosis processes. Journal of Membrane Science. 2010;**355**:36-44

[90] Wang KY, Ong RC, Chung T-S. Double-skinned forward osmosis membranes for reducing internal concentration polarization within the porous sublayer. Industrial & Engineering Chemistry Research. 2010;**49**:4824-4831

[91] Zhang S, Wang KY, Chung T-S, Jean YC, Chen H. Molecular design of the cellulose ester-based forward osmosis membranes for desalination. Chemical Engineering Science. 2011;**66**:2008-2018

[92] Tang CY, She Q, Lay WCL, Wang R, Field R, Fane AG. Modeling double skinned FO membranes. Desalination. 2014;**283**:178-186

[93] Sairam M, Sereewatthanawut E, Li K, Bismarck A, Livingston AG. Method for the preparation of cellulose acetate flat sheet composite membranes for forward osmosis–desalination using MgSO$_4$ draw solution. Desalination. 2011;**273**:299-307

[94] Vos KD, Burris FO, Riley RL. Kinetic study of the hydrolysis of cellulose acetate in the pH range of 2–10. Journal of Applied Polymer Science. 1966;**10**:825-832

[95] Merten U. Flow relationships in reverse osmosis. Industrial & Engineering Chemistry Fundamentals. 1963;**2**:229-232

[96] Widjojo N, Chung T-S, Weber M, Maletzko C, Warzelhan V. The role of sulphonated polymer and macrovoid-free structure in the support layer for thin-film composite (TFC) forward osmosis (FO) membranes. Journal of Membrane Science. 2011;**383**:214-223

[97] McCutcheon JR, Elimelech M. Influence of membrane support layer hydrophobicity on water flux in osmotically driven membrane processes. Journal of Membrane Science. 2008;**318**:458-466

[98] Yu Y, Seo S, Kim I-C, Lee S. Nanoporous polyethersulfone (PES) membrane with enhanced flux applied in forward osmosis process. Journal of Membrane Science. 2011;**375**:63-68

[99] Song X, Liu Z, Sun DD. Nano gives the answer: Breaking the bottleneck of internal concentration polarization with a nanofiber composite forward osmosis membrane for a high water production rate. Advanced Materials. 2015;**23**:3256-3260

[100] Bui N, Lind ML, Hoek EMV, McCutcheon JR. Electrospun nanofiber supported thin film composite membranes for engineered osmosis. Journal of Membrane Science. 2011:385, 10-386, 19

[101] Fayyazi F, Feijani E, Mahdavi H. Chemically modified polysulfone membrane containing palladium nanoparticles: Preparation, characterization and application as an efficient catalytic membrane for Suzuki reaction. Chemical Engineering Science. 2015;**134**:549-554

[102] Setiawan L, Wang R, Li K, Fane AG. Fabrication of novel poly(amide–imide) forward osmosis hollow fiber membranes with a positively charged nanofiltration-like selective layer. Journal of Membrane Science. 2011;**369**:196-205

[103] Saren Q, Qiu CQ, Tang CY. Synthesis and characterization of novel forward osmosis membranes based on layer-by-layer assembly. Environmental Science & Technology. 2011;**45**:5201-5208

[104] Qiu C, Qi S, Tang CY. Synthesis of high flux forward osmosis membranes by chemically crosslinked layer-by-layer polyelectrolytes. Journal of Membrane Science. 2011;**381**:74-80

[105] Hong SU, Bruening ML. Separation of amino acid mixtures using multilayer polyelectrolyte nanofiltration membranes. Journal of Membrane Science. 2006;**280**:1-5

[106] Hong SU, Ouyang L, Bruening ML. Recovery of phosphate using multilayer polyelectrolyte nanofiltration membranes. Journal of Membrane Science. 2009;**327**:2-5

[107] Bruening ML, Dotzauer DM, Jain P, Ouyang L, Baker GL. Creation of functional membranes using polyelectrolyte multilayers and polymer brushes. Langmuir. 2008;**24**:7663-7673

[108] Stanton BW, Harris JJ, Miller MD, Bruening ML. Ultrathin, multilayered polyelectrolyte films as nanofiltration membranes. Langmuir. 2013;**19**:7038-7042

[109] Malaisamy R, Bruening ML. High-flux nanofiltration membranes prepared by adsorption of multilayer polyelectrolyte membranes on polymeric supports. Langmuir. 2005;**21**:10587-10592

[110] Zhang G, Yan H, Ji S, Liu Z. Self-assembly of polyelectrolyte multilayer pervaporation membranes by a dynamic layer-by-layer technique on a hydrolyzed polyacrylonitrile ultrafiltration membrane. Journal of Membrane Science. 2007;**292**:1-8

[111] Jin W, Toutianoush A, Tieke B. Use of polyelectrolyte layer-by-layer assemblies as nanofiltration and reverse osmosis membranes. Langmuir. 2003;**19**:2550-2553

[112] Lee H, Lee Y, Statz AR, Rho J, Park TG, Messersmith PB. Substrateindependent layer-by-layer assembly by using mussel-adhesive-inspired polymers. Advanced Materials. 2008;**20**:1619-1623

[113] Akbari A, Desclaux S, Rouch JC, Aptel P, Remigy JC. New UV-photografted nanofiltration membranes for the treatment of colored textile dye effluents. Journal of Membrane Science. 2006;**286**:342-350

[114] Akbari A, Desclaux S, Rouch JC, Remigy JC. Application of nanofiltration hollow fibre membranes, developed by photografting, to treatment of anionic dye solutions. Journal of Membrane Science. 2007;**297**:243-252

[115] Li X-L, Zhu L-P, Xu Y-Y, Yi Z, Zhu B-K. A novel positively charged nanofiltration membrane prepared from N,N-dimethylaminoethyl methacrylate by quaternization cross-linking. Journal of Membrane Science. 2011;**374**:33-42

[116] Deng H, Xu Y, Chen Q, Wei X, Zhu B. High flux positively charged nanofiltration membranes prepared by UV-initiated graft polymerization of methacrylatoethyl trimethyl ammonium chloride (DMC) onto polysulfone membranes. Journal of Membrane Science. 2011;**366**:363-372

[117] Miao J, Chen G-H, Gao C-J. A novel kind of amphoteric composite nanofiltration membrane prepared from sulfated chitosan (SCS). Desalination. 2005;**181**:173-183

[118] He T, Frank M, Mulder MHV, Wessling M. Preparation and characterization of nanofiltration membranes by coating polyethersulfone hollow fibers with sulfonated poly (ether ether ketone) (SPEEK). Journal of Membrane Science. 2008;**307**:62-72

[119] Achilli A, Cath TY, Childress AE. Selection of inorganic-based draw solutions for forward osmosis applications. Journal of Membrane Science. 2010;**364**:233-241

[120] Kim T-W, Kim Y, Yun C, Jang H, Kim W, Park S. Systematic approach for draw solute selection and optimal system design for forward osmosis desalination. Desalination. 2012;**284**:253-260

[121] Yen SK, Mehnas Haja F, Su NM, Wang KY, Chung T-S. Study of draw solutes using 2-methylimidazole-based compounds in forward osmosis. Journal of Membrane Science. 2010;**364**:242-252

[122] Ling MM, Chung T-S. Desalination process using super hydrophilic nanoparticles via forward osmosis integrated with ultrafiltration regeneration. Desalination. 2011;**278**:194-202

[123] Ling MM, Wang KY, Chung T-S. Highly water-soluble magnetic nanoparticles as novel draw solutes in forward osmosis for water reuse. Industrial & Engineering Chemistry Research. 2010;**49**:5869-5876

[124] Ge Q, Su J, Chung T-S, Amy G. Hydrophilic superparamagnetic nanoparticles: Synthesis, characterization, and performance in forward osmosis processes. Industrial & Engineering Chemistry Research. 2011;**50**:382-388

[125] Ling MM, Chung T-S, Lu X. Facile synthesis of thermosensitive magnetic nanoparticles as smart draw solutes in forward osmosis. Chemical Communications. 2011;**47**:10788-10790

[126] Li D, Zhang X, Yao J, Simon GP, Wang H. Stimuli-responsive polymer hydrogels as a new class of draw agent for forward osmosis desalination. Chemical Communications. 2011;**47**:1710-1712

[127] Li D, Zhang X, Yao J, Zeng Y, Simon GP, Wang H. Composite polymer hydrogels as draw agents in forward osmosis and solar dewatering. Soft Matter. 2011;**7**:10048-10056

[128] Liu Z, Bai H, Lee J, Sun DD. A low-energy forward osmosis process to produce drinking water. Energy & Environmental Science. 2011;**4**:2582-2585