Forward osmosis: Where are we now?

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HIGHLIGHTS
• We present a critical review of the current state of forward osmosis (FO).
• We analyze the energy efficiency of FO and emphasize relevant applications.
• We discuss the key required membrane properties for FO and future implications.
• We highlight fouling reversibility of FO and relevant benefits and applications.
• We discuss applications where FO outperforms current technologies.

ABSTRACT
Forward osmosis (FO) has been extensively investigated in the past decade. Despite significant advancements in our understanding of the FO process, questions and challenges remain regarding the energy efficiency and current state of the technology. Here, we critically review several key aspects of the FO process, focusing on energy efficiency, membrane properties, draw solutes, fouling reversibility, and effective applications of this emerging technology. We analyze the energy efficiency of the process, dispel the common misguided notion that FO is a low energy process, and highlight the potential use of low-cost energy sources. We address the key necessary membrane properties for FO, stressing the importance of the structural parameter, reverse solute flux selectivity, and the constraints imposed by the permeability–selectivity tradeoff. We then dispel the notion that draw solution regeneration can use negligible energy, highlighting the beneficial qualities of small inorganic and thermolytic salts as draw solutes. We further discuss the fouling propensity of FO, emphasizing the fouling reversibility of FO compared to reverse osmosis (RO) and the prospects of FO in treating high fouling potential feed waters. Lastly, we discuss applications where FO outperforms other desalination technologies and emphasize that the FO process is not intended to replace RO, but rather is to be used to process feed waters that cannot be treated by RO.

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1. Introduction

Forward osmosis (FO), an emerging separation/desalination process, has received increased attention in the past decade in both academic research and industrial development [1-2]. In FO, a semipermeable membrane is placed between two solutions of different concentrations: a concentrated draw solution and a more dilute feed solution. By using the osmotic pressure difference to drive the permeation of water across the membrane, FO can address several shortcomings of hydraulic pressure-driven membrane processes, such as reverse osmosis (RO).

Early studies focused on various potential applications of FO in the food, water, and energy sectors [1]. The introduction of the ammonia–carbon dioxide FO process in 2005 as a potential desalination process that utilizes low-grade thermal energy [3] has stimulated academic and industrial interest in FO, which resulted in a dramatic increase in the number of research articles and patents in subsequent years [2-4]. These studies on FO involved membrane development [5-8], mass transfer analysis [9,10], membrane characterization [11,12], and introduction and characterization of new draw solutions [13-16]. Concurrently, conceptual and bench-scale studies on various potential applications of FO have been published, including the use of FO coupled with a draw solution separation/regeneration stage [21-23], FO in osmotic dilution processes [23,24], and various hybrid systems incorporating FO [25-28].

Despite the recent advancements in FO, there remain several challenges to overcome for successful implementation of the technology. Moreover, confusion exists regarding the energy consumption by the FO process, triggered by misguided studies defining FO as a “low energy process.” Other studies present FO as an alternative to RO, a robust pressure-driven membrane desalination process. While several review articles on FO have been published recently [2,4], none has critically addressed the energy efficiency of FO, the viability of the technology, and the applications where FO has clear advantages over conventional desalination processes. A review that analyzes these key points as well as other enabling aspects of FO is crucially needed.

In this review article, we critically discuss the energy efficiency, membrane performance, optimal draw solutes, and suitable applications of FO. Specifically, we address the following key questions: Is FO a low energy process? What are the key required membrane properties for FO? Is finding a magic draw solution the Holy Grail in FO? Is FO a low fouling process? Where does FO outperform other desalination technologies? Addressing these questions and understanding the limits of FO will provide vital information to further advance the technology and expand the range of its applications.

2. Is forward osmosis a low energy process?

2.1. No “free lunch” for FO

The fact that water in FO permeates spontaneously through a semipermeable membrane does not mean that FO is more energy efficient as a separation process than other membrane processes. In fact, FO is not only a separation process, but is simultaneously a separation and mixing process. The water molecules that transport across the membrane from a salty feed solution mix with the draw solution to reduce its chemical potential. In order to obtain fresh water as a product, further separation of the diluted draw solution is required subsequent to the FO process. Based on thermodynamic principles and practical kinetic requirements, the theoretical minimal energy for desalination with FO is always higher than that without FO. In other words, using FO cannot reduce the minimum energy of separation.

This general conclusion regarding the FO desalination energy has fundamental underlying thermodynamic rationales. In an isothermal separation process, energy is required to reduce the entropy of the system [29,30]. However, the spontaneity of the FO process implies that entropy is generated [31] and that the system entropy of the intermediate state (i.e., when the draw solution is diluted and feed solution is concentrated) is higher than that of the initial state. Regardless of the process used, separation of the feed solution to the same degree should result in identical system entropy in the final state. Therefore, the minimum energy for the post-FO separation stage, which is required to reduce the system entropy from the intermediate to the final state, is obviously higher than the minimum energy for a standalone separation, which is required to reduce the system entropy only from the initial to the final state. To further elucidate this qualitative argument, an analysis is conducted in Section 2.2 to compare the minimum energy requirement of a standalone RO process to that of an FO–RO hybrid process.

2.2. FO–RO consumes more electric energy than RO alone

For comparison of overall energy consumption between an RO process and an FO–RO process, it is a reasonable approximation to consider only the energy required for the RO separation. This approximation assumes that the energy requirement in RO for generating the hydraulic pressure to overcome the osmotic pressure difference between the concentrated and dilute solutions dominates the overall energy consumption, rendering the energy for flow circulation and other practical considerations in the RO and FO systems relatively insignificant [29]. Therefore, comparing the energy consumption of a standalone RO system and a hybrid FO–RO system can be approximately reduced to the comparison between the energy consumption of the RO stages in these respective systems.

We now compare the energy consumption between a standalone RO process (denoted as RO1 in Fig. 1A) and an FO–RO process (denoted as FO–RO2 in Fig. 1A). In the RO1 process, the feed solution is separated into the brine solution (the red block in Fig. 1A) and the permeate solution (the blue block). In the FO process, water molecules in the feed solution migrate across the semipermeable membrane to mix with the draw solution (the green block), resulting in the diluted draw solution (the blue and green composite block) and the same brine solution as in RO1. The diluted draw solution is then separated by the RO2 process to produce a permeate solution of the same volume as in the RO1 process, and a concentrated draw solution of the same volume and
concentration as the initial draw solution. For the RO₁ and the FO–RO₂ processes to achieve the same separation, the osmotic pressure of the brine solution in the RO₁ process, \( \pi_{\text{RO₁,brine}} \), and the osmotic pressure of the brine solution in the FO process (i.e., concentrate of the FO feed solution), \( \pi_{\text{FO,feed}} \), have to be equal:

\[
\pi_{\text{RO₁,brine}} = \pi_{\text{FO,feed}}. \tag{1}
\]

In an RO process, the minimum specific energy (i.e., energy consumed per unit volume of product water) to achieve a certain recovery is equal to the brine osmotic pressure, \( \pi_{\text{RO₁,brine}} \) [32]. In an ideal FO–RO system with perfect salt rejection, the draw solution is conserved, and the brine of the RO₂ process is simply the initial draw solution in the FO process, \( \pi_{\text{RO₂,brine}} = \pi_{\text{FO,draw}} \), such that:

\[
\pi_{\text{RO₂,brine}} = \pi_{\text{FO,draw}}. \tag{2}
\]

The condition for net driving force in FO dictates that the feed osmotic pressure must always be lower than the draw solution osmotic pressure in either co-current (Fig. 1B) or counter-current (Fig. 1C) membrane module configuration. The corollary inequality of the net driving force condition, \( \pi_{\text{FO,feed}} < \pi_{\text{FO,draw}} \), when coupled with the conditions given by Eqs. (1) and (2), suggests that:

\[
\pi_{\text{RO₁,feed}} > \pi_{\text{RO₂,feed}}. \tag{3}
\]

The inequality in Eq. (3) indicates that the minimum specific energy for RO₂ is higher than that in RO₁ in order for the FO–RO₂ and the RO₁ processes to achieve the same recovery. Therefore, a standalone RO process is more energy efficient than an FO–RO hybrid process.

Although only the theoretical minimum energy of separation is compared in our analysis, incorporating other practical considerations, such as concentration polarization and excess pressure, will unlikely change the relative energetic advantage of RO over FO–RO. Our analysis, although highly simplified, leads to the same conclusion obtained from a detailed comparison between an RO and an FO–RO process when considering other practical factors, such as energy for cross-flow circulation and the energy requirement for ultrafiltration pretreatment when RO is used without FO [33]. The itemized assessment of energy consumption for a detailed comparison of RO and FO–RO processes illustrated that the energy cost of RO dominates the total energy cost for both RO and FO–RO processes, which justifies the assumption in our simplified analysis. We also note that an intrinsic trade-off exists between the energy efficiency of an FO–RO process, which is quantified by specific energy, and the mass transfer kinetics in FO, which can be characterized by average water flux and are controlled by the overall osmotic pressure driving force. The enhanced mass transfer kinetics in FO must be achieved at the cost of higher energy consumption in the subsequent RO process.

### 2.3. Opportunities in leveraging low-cost thermal energy

Although incorporation of FO in a separation process cannot reduce the minimum energy of separation compared to a standalone separation process (RO or other methods), FO hybrid systems may still provide energy cost savings, if alternative low-cost thermal energy can be used to power the post-FO separation process. For example, a thermolytic solution of ammonia–carbon dioxide has been proposed as a draw solution in FO, in which low-grade heat energy can be exploited to regenerate the draw solution by preferentially removing the solutes that are significantly more volatile than water [3,34]. Alternatively, FO can also be coupled with membrane distillation (MD) to desalinate waters that are challenging for a standalone MD process. In a hybrid FO–MD system, FO is applied to mitigate organic fouling and/or mineral scaling that are detrimental to the MD process, whereas the MD process separates the water and regenerates the draw solution using low-grade heat [27]. Further discussion on FO hybrid processes that utilize low-cost thermal energy for separation is provided in Section 6.2.
3. What are the key required membrane properties for FO?

The ideal FO membrane achieves high water flux, completely rejects feed and draw solutes, and is mechanically robust. Membrane properties for the support and active layers are critical for realizing these aims. In this section, we highlight two particularly important membrane design goals: (i) minimizing the structural parameter of the support layer to mitigate mass transfer limitations and increase water flux, and (ii) maximizing the reverse solute flux selectivity of the active layer to limit loss of draw solute.

3.1. Low structural parameter to maximize water flux

The phenomenon of internal concentration polarization (ICP) can have pronounced performance-limiting effects in FO [35–37]. ICP arises due to the porous support layer serving as an unsterilized diffusive boundary layer [6]. As water permeates the membrane active layer from the feed solution and dilutes the draw solution at the interface between the active layer and the porous support, the driving force for permeation decreases. This draw solution dilution is partially counteracted by draw solute back-diffusion from the bulk draw solution into the support. However, the result is a concentration profile in which the draw solute concentration at the active layer interface is lower than in the bulk, reducing the osmotic pressure difference across the active layer and therefore reducing water flux.

To minimize ICP and maximize water flux, resistance to draw solute diffusion through the support layer must be minimized. FO membrane support layer properties are crucial in this aim. The ideal support layer is thin, highly porous, and minimally tortuous, all of which minimize the diffusion path and enhance back-diffusion of draw solute. Membrane support chemistry, most notably hydrophilicity, can also play an important role in the severity of ICP. Hydrophobic support materials, such as the commonly-used polysulfone and polyester, do not fully wet when exposed to water, decreasing the effective porosity of the support layer [38]. These support layer properties, together with the solute diffusivity, comprise the resistance to solute diffusion, \(K\), which can be expressed as the reciprocal of a thin-film mass transfer coefficient [39]:

\[
K = \frac{t_s \tau}{D_{\text{eff}}} \tag{4}
\]

where \(D\) is the solute diffusion coefficient, \(t_s\) is the support layer thickness, \(\tau\) is the support layer tortuosity, and \(D_{\text{eff}}\) is the effective support layer porosity. The membrane properties can be grouped to define the structural parameter \(S\) [5,40]:

\[
S = KD = \frac{t_s \tau}{D_{\text{eff}}} \tag{5}
\]

Since its introduction, the structural parameter has provided a simple benchmark for the design and comparison of FO membranes.

Until recently, studies of FO predominantly utilized polyamide-based thin film composite (TFC) RO membranes or the commercial asymmetric cellulose triacetate (CTA) FO membrane by Hydration Technology Innovations (HTI, Albany, OR) [38,41]. TFC membranes are the gold standard for RO because of their high water permeability, high solute selectivity, and chemical and physical stability [5,42,43]. However, use of TFC-RO membranes in the FO process resulted in very low water fluxes due to their dense support layers and thick fabric backing [5]. These extensive support layers are crucial for maintaining mechanical integrity under the high hydraulic pressures used in RO, but the same support is unnecessary in FO and leads to very large structural parameters of nearly 10,000 \(\mu \text{m}\) [5]. With its relatively low structural parameter of 500 \(\mu \text{m}\), the HTI-CTA FO membrane obtained the best performance in FO, despite having lower permeability and selectivity than polyamide TFC-RO membranes [5,9–11,41].

Recent research has led to dramatic decreases in the structural parameter [5,6,8,44–48]. A major advance was the formation of robust TFC membranes with phase inversion-formed supports that were specifically tailored to minimize the structural parameter. Through proper choice of solvents for phase inversion and use of a thin fabric support, hand-cast TFC membranes were fabricated with an average \(S\) of 492 \(\mu \text{m}\), in comparison with 9583 \(\mu \text{m}\) for a commercial TFC-RO membrane (SW30, Dow Chemical Company, Midland, MI) [5]. Subsequent studies reported structural parameter values of less than 100 \(\mu \text{m}\) for asymmetric membranes formed through phase inversion [8] and for TFC membranes formed through interfacial polymerization of a polyamide film directly on an electrospun polyethersulfone support [46]. However, the properties that contribute to a low structural parameter can also lead to mechanical fragility, and it remains to be seen whether ultra-low \(S\)-value membranes will be robust enough to withstand the hydrodynamic stresses inherent in FO operation.

FO membrane research has rapidly translated into commercially-available, high-flux, TFC-FO membrane modules (Oasys Water Inc., Boston, MA: HTI), most of which have structural parameters less than 500 \(\mu \text{m}\) [5,9–11,47–49], FO membrane development and its impact on water flux performance are demonstrated in Fig. 2, which models flux based on published water permeability, \(A\), salt permeability, \(B\), and \(S\)-values for commercial membranes considering a 1 M sodium chloride draw solution and deionized water feed solution. The water flux model is based on the film theory [10]:

\[
J_w = A \frac{\frac{\pi_0}{w} \exp\left(-\frac{J_{\text{os}} S}{B}\right) - \pi_f \exp\left(-\frac{J_{\text{os}} S}{B}\right)}{1 + \frac{B}{J_w} \left(\frac{\pi_0}{w} \exp\left(-\frac{J_{\text{os}} S}{B}\right) - \exp\left(-\frac{J_{\text{os}} S}{B}\right)\right)} \tag{6}
\]

where \(J_w\) is the volumetric water flux, \(n_0\) is the bulk draw solution osmotic pressure, \(n_f\) is the bulk feed solution osmotic pressure, and \(k_i\) is the feed-side mass transfer coefficient. For the conditions modeled in Fig. 2, recently commercialized TFC-FO membranes can achieve FO
water fluxes that are 10–20 times higher than a TFC-RO membrane and are twice the water flux of asymmetric CTA FO membranes. However, despite the achievement of relatively high water fluxes, opportunities for improvement remain. With improved mechanical integrity, practical FO membranes with ultra-low S-values may be attained. In addition, all of the asymmetric CTA membranes and some of the TFC-FO membranes have relatively low selectivity compared to state-of-the-art seawater TFC-RO membranes [42,50]. A highly selective FO membrane with a low structural parameter has yet to be developed.

3.2. High reverse solute flux selectivity (RSFS) to minimize loss of draw solute

Loss of draw solute during FO operation occurs via reverse solute flux, which refers to the back permeation of draw solutes from the bulk draw solution through the membrane active layer and into the feed. The ability of an FO membrane to minimize loss of draw solute is captured by the reverse solute flux selectivity (RSFS). RSFS represents the volume of water produced per mass of draw solute lost, and it follows a simple, experimentally-verified relationship based on the van’t Hoff equation [9,10]:

\[ \text{RSFS} = \frac{J_w}{J_f} = \frac{A}{B} n R_\text{g} T \]

where \( J_s \) is reverse mass flux of draw solute, \( n \) is the number of dissolved species created by the draw solute (e.g., 2 for NaCl), \( R_\text{g} \) is the ideal gas constant, and \( T \) is the absolute temperature. The RSFS is independent of support layer properties (i.e., structural parameter) and bulk draw solution concentration, and it is solely dependent on the permselectivity of the active layer, namely \( A \) and \( B \). To maximize RSFS, the \( A/B \) ratio must be maximized.

Maximizing RSFS is critically important because the loss of draw solute (i) is economically unfavorable, (ii) can lead to enhanced fouling and scaling in the feed solution [52,53], and (iii) for some draw solutes, can have negative impacts upon release to the environment (e.g., ammonia [54]). Using a highly selective membrane would minimize the detrimental effects of draw solute loss and would allow for the selection of small, fast-diffusing draw solutes, such as sodium chloride, magnesium chloride, and ammonia–carbon dioxide [1,55]. As discussed further in Section 4, these solutes are ideal for minimizing ICP and creating high osmotic pressures. However, they are prone to relatively high reverse solute fluxes due to their small size [17,55]. Ammonia–carbon dioxide, which holds great promise as a draw solute due to its ability to be thermally evaporated for facile draw solution regeneration [3,48], suffers from particularly high membrane solute permeability. Ammonia–carbon dioxide specific reverse salt fluxes are nearly an order of magnitude higher than sodium chloride through asymmetric CTA membranes and are three times higher than sodium chloride through TFC-FO membranes [55,56]. Development of highly selective membranes could decrease reverse solute flux to low levels, even for ammonia–carbon dioxide.

High \( A/B \) ratios also increase forward solute flux selectivity, thus decreasing forward permeation of undesired feed solutes. This is crucial in all FO applications, including hybrid systems such as FO–RO and FO-distillation. In these systems, solutes that diffuse into the draw solution can accumulate over time, eventually leading to precipitation in the draw solution or solute permeation into the final product water [55,57]. Maintaining high selectivity in the first barrier (FO) mitigates these issues.

While achieving high \( A/B \) ratios is critical, the fabrication of highly selective membranes is constrained by the water permeability–solute selectivity tradeoff [58]. This tradeoff has been postulated to be an intrinsic property of water transport through polymeric membranes, including polyamide-based TFC membranes. Recent analyses led to the following empirical relationship [58,59]:

\[ B = \frac{l^2}{\lambda} \left( \frac{R_T}{M_w} \right) \frac{1}{A^3} \]

where \( l \) is the thickness of the active layer, \( \lambda \) is a fitting parameter, and \( M_w \) is the solute molecular weight. This empirical relationship matches closely with the experimental data shown in Fig. 3 [59], and the cubic relationship of \( B \) with \( A \) immediately demonstrates that increasing water permeability comes with rapidly decreasing selectivity. This relationship provides tremendous insight for the fabrication of selective TFC membranes: to achieve TFC membranes with high RSFS, water permeability must be sacrificed. Ultimately, we anticipate that the best-performing TFC-FO membrane will have modest water permeability, high selectivity, and an ultra-low structural parameter. Such a membrane would achieve high water fluxes with minimal loss of draw solute.

4. Is finding a magic draw solution the Holy Grail in FO?

Similar to FO membrane properties, the choice of draw solute can have a large impact on the performance and viability of the FO process. The ideal draw solute is inexpensive, stable, non-toxic, highly soluble, and has a molecular size that is large enough to limit reverse draw solute flux through the FO membrane active layer, yet small enough to be highly mobile and mitigate ICP [17,20]. In addition, draw solutes should be able to generate high osmotic pressures and be completely regenerated using simple, efficient techniques. Due to these numerous and sometimes conflicting requirements and the large number of potential solutes that could be used, draw solutes with strikingly different properties have been tested for FO [17–20,60–64]. It is clear that a focused and updated discussion of desired draw solute properties is needed. To that end, this section (i) dispels the notion that draw solutes can be regenerated using negligible energy, (ii) discusses the simple solutes that have emerged as optimal FO draw solutes, and (iii) assesses where draw solute innovation would be most impactful.

4.1. The choice of draw solute cannot decrease the minimum energy of separation

Recent research [17–20,60–64] has led to numerous ideas for draw solutes, including polyelectrolytes [18], hydrophilic magnetic
Fig. 4. Relationship between osmotic pressure and viscosity for different draw solutions. The solid curves represent draw solutions made of simple small ions. The red open triangles represent a draw solution with polyacrylic acid (MW = 1800 Da) as the draw solute [18]. The black open hexagons indicate a draw solution with a hexavalent phosphazen salt, hexa(4-ethylcarboxylatophenoxyl)phosphazene, as the draw solute [64]. All other symbols are representative of draw solutions using different tertiary amines and dissolved carbon dioxide as switchable polarity solvents for draw solutes [19,65]. The data for the solid curves were simulated using OLI Stream Analyzer (OLI Systems, Inc. Morris Plains, NJ) at a temperature of 25 °C. Due to complexities in modeling the ammonia–carbon dioxide system (i.e., obtaining a continuous curve), aqueous solutions of ammonia carbamate were modeled to represent the osmotic pressure and viscosity relationship of ammonia–carbon dioxide. All other data were collected from the corresponding literature. The vertical dotted line indicates the viscosity of pure water at 25 °C. The dash-dot lines show the representative osmotic pressures of three feed solutions: shale gas produced water (64.8 bar), seawater (27 bar), and brackish water (7.7 bar).

nanoparticles [63], hydrogels [61,62], and switchable polarity solvents [19,65]. A major driver in these studies is finding a draw solute that can decrease the energy of draw solution regeneration. However, potential energy savings from novel draw solutes are limited. As demonstrated in Section 2.1, the energy requirement of an FO process (with any draw solution regeneration scheme) cannot be less than the minimum energy of separation. In addition, the energy consumption for RO in state-of-the-art processes already approaches this minimum energy [32]. Therefore, it is extremely unlikely that any draw solution regeneration technology will prove more energetically favorable than RO, and when it can be applied, RO will remain the optimal draw solution regeneration technology. Novel draw solution regeneration technologies seeking gains in energy efficiency must focus on high osmotic pressure applications for which RO cannot be used.

A simple example to further illustrate energetic limitations during draw solution regeneration is the use of polyelectrolytes as a draw solute. Polyelectrolyte draw solution regeneration was performed by ultrafiltration (UF) [18], which is typically a low-pressure and low-energy process in comparison with RO. However, if the polyelectrolyte creates osmotic pressure to drive FO, and is completely regenerated by UF, then the osmotic pressure of the UF brine, \( \pi_{\text{UF},\text{brine}} \), must be equal to the inlet osmotic pressure of the FO draw solution:

\[
\pi_{\text{UF},\text{brine}} = \pi_{\text{FO,draw}}. \tag{9}
\]

Comparison of Eqs. (9) and (2) results in:

\[
\pi_{\text{UF},\text{brine}} = \pi_{\text{UF},\text{brine}} - \pi_{\text{FO,draw}}. \tag{10}
\]

The UF minimum operating hydraulic pressure in an FO–UF process is the same as that for RO in an FO–RO process, contrary to the usual expectation of UF being low-pressure and low-energy. Thus, the minimum energy requirements for FO–UF and FO–RO are the same, and using a large polyelectrolyte does not save any energy compared to FO–RO or standalone RO. This conclusion can be generalized for the use of any pressure-driven membrane process as the draw solution regeneration step.

4.2. Simple dissolved salts remain the optimal draw solutes

Despite the extensive research efforts into novel draw solutes, simple dissolved inorganic and thermolytic salts (e.g., sodium chloride, magnesium chloride, and ammonia–carbon dioxide) remain the most widely-used, and arguably the most effective. These small inorganic solutes have two distinct and important advantages: (i) their ability to generate high osmotic pressures at low solution viscosities, and (ii) their high diffusivities that mitigate the flux-limiting effect of ICP.

High osmotic pressures are critical for FO, as the most impactful applications are for high salinity feeds for which RO cannot be used. In general, the osmotic pressure of a solution, \( \pi \), can be related to the mass concentration of the solute, \( c \), by the following equation:

\[
\pi = cR_T \left( \frac{1}{M} A_2 c + A_3 c^2 + \ldots \right) \tag{11}
\]

where \( M \) is the molecular weight of the solute, and \( A_2 \) and \( A_3 \) are the second and third virial coefficients that capture the solute–solvent interactions [66]. For estimating the order of magnitude, the osmotic pressure is inversely proportional to the molecular weight of the solute. Thus, as shown in Fig. 4, low molecular weight inorganic solutes easily generate osmotic pressures exceeding 100 bar at viscosities near that of pure water (i.e., 1 cP). In comparison, nanoparticles and polymer based draw solutes, which are extremely large compared to dissolved salts, are unable to reach an appreciable osmotic pressure without using very high solute mass concentrations that lead to unacceptably high
viscosity. Low solution viscosity is strongly desired to minimize hydraulic pressure losses across the membrane channel.

The second advantage of small inorganic solutes lies in their relatively high diffusivities, which decrease ICP and lead to higher water flux. The importance of limiting ICP was discussed extensively in Section 3.1 as related to membrane properties. As can be seen from Eq. (6) and in Fig. 5, the draw solute diffusivity is critical, having an exponential effect on the water flux. Even relatively small variations among the diffusivities of inorganic salts can have pronounced effect on water flux [17]. Compared to simple inorganic salts, some of the recently proposed nanoparticle and polyelectrolyte draw solutes are orders of magnitude larger, with characteristic sizes varying from several nanometers to tens of nanometers [18,63]. Fig. 5A illustrates that these extremely large draw solutes suffer from significant ICP and cannot generate high water fluxes, even when using membranes with low structural parameters. Thus, their application to the treatment of realistic feed solutions, such as brackish water, seawater, or shale gas produced water, is limited as shown in Fig. 5B.

When choosing draw solutes, there is a tradeoff between small solute size to limit ICP and large solute size to decrease reverse draw solute flux. Indeed, one of the main advantages of polymer and nanoparticle-based draw solutions is essentially eliminating reverse draw solute flux. However, with the recent commercialization of TFC-FO membranes, which have much higher RSFS than CTA membranes [10,60], reverse draw solute flux for small inorganic solutes has been greatly reduced. This trend will continue with the expected commercialization of more highly selective TFC-FO membranes, further decreasing the need for exploring large draw solutes, and further promoting the use of small, highly-mobile solutes.

4.3. Some opportunity remains for draw solute innovation

While inorganic and thermolytic salts should adequately serve most FO applications, marginal benefits may be obtained using specialized draw solutes for niche applications. Some FO draw solutes are strategically chosen to serve as the final product, most notably concentrated fertilizer for agricultural uses [68] and concentrated nutrients in commercial hydration bags sold by HTI. Large draw solutes may find application in low osmotic pressure applications for which even minimal reverse solute flux is unacceptable (e.g., liquid food processing and pharmaceutical concentration). However, for these low-pressure applications, conventional pressure-driven membrane processes will likely prove much more efficient and outperform FO.

More significant opportunity lies in innovative high osmotic pressure draw solutes, such as switchable polarity solutes [19,65], or novel thermolytic draw solutes, such as trimethylamine (TMA)—carbon dioxide [60]. Switchable polarity solutes considered thus far for FO have all been tertiary amines that phase transition from water-miscible to water-immiscible depending on the presence of carbon dioxide [19,65]. Studies of switchable polarity solutes are still preliminary, and energy requirements (i.e. heat input) have yet to be analyzed or disclosed. However, some of the solutes studied have demonstrated high osmotic pressures at reasonable viscosities (Fig. 4), and the ability to separate the draw solute from water in the liquid phase may prove beneficial. Reduced reverse solute flux and draw solution regeneration energy are the main advantages for using TMA—carbon dioxide as a draw solute compared to ammonia—carbon dioxide [60]. While their potential is considerable, switchable polarity solvents and TMA have toxicity concerns that will likely limit their use to industrial applications [19,60].

5. Is FO a low fouling process?

5.1. FO fouling is highly reversible

Fouling involves the deposition and adsorption of feed water constituents, such as organic and inorganic compounds, colloidal particles, and microbes, to the membrane surface. As water permeates the
Recent studies indicate that FO is a low fouling process because the foulant layer formed in FO is structurally different from fouling in pressure-driven membrane processes [12,26,52,71]. The structure of the cake layer formed during RO is densely compacted, whereas the cake layer in FO is thicker but much less compact [52]. Because of the loosely-packed fouling layer, FO can recover as much as 80–100% of the initial water flux through periodic rinses to clean the membrane surface [71]. In comparison, RO fouling is mostly irreversible without chemical cleaning [52].

Fig. 6A demonstrates how simple rinsing at a high cross-flow velocity with a low ionic strength solution accomplishes complete removal of an alginate fouling layer from an FO membrane [13]. Reversibility is illustrated by the recovery of the water flux after rinsing (blue circles) to the initial water flux prior to fouling (red squares). Such high reversibility is similarly observed with scaling by gypsum [72] and silica [73], and fouling by proteins (Fig. 6B).

Membrane fouling in FO can also be mitigated by introducing hydrodynamic shear forces near the membrane surface to prevent foulant accumulation. Spacers placed alongside the membrane induce mixing and have been shown to prevent the attachment of foulants [74,75]. Other hydrodynamic strategies that mitigate fouling in FO include operating at a higher cross-flow velocity [52], use of flow pulsation [74], and introducing air bubbles [13]. However, these hydrodynamic approaches will increase the energy consumption of the system, which is a consideration for any fouling mitigation strategy.

High reversibility of fouling in FO enables the treatment of raw feed waters with high fouling potential, such as those heavily laden with organic material (e.g., natural and effluent organic matter) [76]. Several studies have demonstrated that a steady water flux is maintained during wastewater treatment [74,76] and sludge dewatering [77] by FO. Nonetheless, fouling is undesirable as it decreases process performance and increases operational costs. Therefore, understanding fouling mechanisms and developing fouling mitigation methods continue to be active areas of research [74].

5.2. Membrane surface properties are important for fouling control

Fouling propensity in FO is dictated by hydrodynamic operation conditions, as discussed in Section 5.1, and also by the affinity between foulants and the membrane surface [32]. As such, a fouling-resistant FO membrane is characterized by an inert surface chemistry to prevent attracting foulants and a smooth surface topography that impedes foulant entrapment.

Membranes utilized thus far in FO have predominantly been either asymmetric cellulose triacetate (CTA) or polyamide thin film composites (TFC). Although organic fouling of FO has been a prevalent topic in recent publications, the vast majority of the fouling studies were conducted with CTA membranes [12–14,26,52,71–82] because no TFC-FO membranes were commercially available until recently. As discussed in Section 3, TFC membranes are the current state-of-the-art for FO because they exhibit a higher water permeability and salt rejection than CTA membranes [5]. Thus, there is a critical need for systematic research on the fouling resistance of TFC-FO membranes, which are inherently prone to fouling because of their high surface roughness, relative hydrophobicity, and the presence of carboxyl groups on the membrane surface [32].

In the few studies on FO fouling that have used TFC membranes, efforts have focused on surface modification of the membrane active layer to mitigate their fouling propensity [15,83–85]. A common method to impart antifouling properties is by grafting hydrophilic polymers, such as poly(ethylene glycol) (PEG), to the membrane active layer to create a polymer brush, which acts as a steric barrier to the adsorption of foulants [15,83,86]. These modified membranes exhibit a significant decrease in water contact angle and reduced foulant–membrane adhesion forces, indicating an increase in fouling resistance. However, this improvement in fouling resistance is achieved at the cost of reduced water permeability that is attributed to the hydraulic resistance of the grafted polymer layer [15,83].

In organic fouling tests, FO membranes modified with PEG demonstrated reduced flux decline due to organic fouling and were capable of recovering 100% of the initial water flux after rinsing, as illustrated in Fig. 7 [83]. As such, surface modification reduces the propensity for membrane fouling and improves the reversibility of fouling, thereby enabling the treatment of high fouling potential feed waters by FO. A review on surface modifications for antifouling membranes can be found elsewhere [87].
Data from Lu et al. [83].

6. Where does FO outperform other desalination technologies?

6.1. FO excels with challenging feed waters

FO must be paired with a draw solution regeneration process for desalination applications, and the most studied of these hybrid FO desalination systems is FO–RO. As discussed in Section 2.2, a hybrid FO–RO system will not consume less energy for separation than an RO system, and FO will likely not replace RO as a standalone process for brackish water or seawater desalination.

While FO is not more energetically favorable than RO for separation, in applications with feed waters that are challenging to treat because of high salinity, high fouling potential, or the presence of specific contaminants, FO can outperform or enhance RO and other desalination technologies. Hybrid FO-distillation systems may be used to treat high-salinity feed waters with osmotic pressures that exceed the working pressure of RO. By employing thermolytic draw solutes, these hybrid systems may use less energy for desalination than standalone thermal technologies. FO can also improve the performance and water quality of conventional membrane and thermal desalination processes when used as pretreatment because of its reduced fouling propensity and ability to remove dissolved constituents. Osmotic dilution may be employed in the treatment of impaired waters to reduce energy consumption and mitigate environmental impacts of treatment. For all FO hybrid systems, the potential benefits and expanded applications that result from incorporating FO are achieved at the cost of the required membrane area and associated materials and equipment for the FO system.

6.2. Hybrid FO systems can desalinate high-salinity feed waters using less energy

Hybrid FO systems will not consume less energy than the comparable standalone desalination technology to achieve the same separation, and this difference in energy consumption is especially evident when operating near the minimum specific energy of separation. However, hybrid FO systems have the potential to consume less total energy than other technologies when desalinating high-salinity feed waters using draw solutions composed of thermolytic salts. To the extent that high-salinity waters also have high organic fouling potential, FO hybrid systems are expected to effectively treat these difficult feed waters because of the fouling resistance of the FO process, as discussed in Section 5.

High-salinity feed waters include brines, such as concentrate from seawater RO [88] or oil and gas produced water [57]. Other high-salinity sources are feed waters that must be treated to very high recovery and corresponding feed water salinity, such as desalination of landfill leachate [89], industrial wastewater [90], and flue gas desulfurization wastewater [91], for which the concentrate is treated as hazardous waste and must be minimized. Brackish water desalination in inland locations, where brine disposal options are limited and costly, is also a source of high-salinity concentrate streams [92]. Zero-liquid discharge treatment schemes are pursued in situations where economic and regulatory limitations make the disposal or discharge of high-salinity water infeasible.

Thermal desalination is required for separation of high-salinity feed waters because the osmotic pressure of the feed water exceeds the operating pressure of RO, which is approximately 70 bar [42,50]. Membrane separation by electrodialysis is also not feasible because the required energy for separation is proportional to the feed water salinity and the required permeate water quality, making desalination of high-salinity solutions uneconomical [93]. Multi-effect distillation (MED), multi-stage flash (MSF), and mechanical vapor compression (MVC) are the technologies commonly used for desalinating high-salinity feed waters. These distillation technologies heat the high-salinity feed solution to vaporize the water, leaving behind salts and other non-volatile feed water contaminants, and they operate far from the minimum specific energy referenced in Section 2.2.

Hybrid FO systems that utilize thermolytic draw solutions may be energetically favorable to distillation technologies for treating high-salinity feed solutions because only relatively small volumes of thermolytic draw solute must be vaporized and recovered from the draw solution, as opposed to vaporizing and recovering the solvent (water) in conventional distillation [57]. The energy required for draw solute recovery is related to the draw solute vapor pressure, and draw solutes with high vapor pressures require less total energy for recovery. By using low-temperature distillation processes for thermolytic draw solute recovery, hybrid FO systems can also leverage low-cost thermal energy sources, such as solar thermal energy, geothermal energy, and...
industrial waste heat, to reduce the energy cost of separation. FO combined with membrane distillation (FO–MD) is one example [27,94].

In FO applications, thermolytic salts with vapor pressures much higher than the vapor pressure of water are desirable as draw solutes because they can be vaporized from the draw solution by a distillation process operating at relatively low temperatures. The most studied thermolytic draw solution is the ammonia–carbon dioxide system [3,95]. In this draw solution, ammonium bicarbonate and ammonium hydroxide salts, which have low molecular weights and high solubility, are dissolved in water to create a concentrated draw solution with high osmotic pressure [41]. Upon moderate heating, the ammonium salts are decomposed into ammonia and carbon dioxide gases, which may be recovered and reconstituted into the draw solution [3,96].

Oasys Water Inc. recently demonstrated the use of a hybrid FO system with an ammonia–carbon dioxide draw solution and a distillation column for draw solute recovery for desalination of high-salinity shale gas produced water [48,97]. The specific energy consumption of the Oasys hybrid FO system was reported to be significantly lower than other thermal distillation methods [48]. Oasys Water also recently announced plans to install a similar system for treatment and concentration of RO brine from treatment of flue gas desulfurization wastewater from a coal-fired power plant. The Oasys hybrid FO-distillation system will be integrated into a zero-liquid discharge facility design as a brine concentrator upstream of a crystallizer [98]. An example schematic of a zero-liquid discharge treatment system incorporating a hybrid FO process with a thermolytic draw solution is shown in Fig. 8.

**Fig. 8.** Schematic of a zero-liquid discharge treatment system incorporating forward osmosis with a thermolytic draw solution. The forward osmosis process serves to further concentrate the brine from the reverse osmosis process before the concentrated brine is treated in a crystallizer.

6.3. FO pretreatment can improve the performance of conventional desalination processes

Conventional desalination technologies include both membrane-based separation processes, such as RO, nanofiltration (NF), and electrodialysis, and thermal desalination technologies like MED, MSF, and MVC. Feed water pretreatment is critical to protect the physical equipment of these conventional processes from damage by feed water constituents and to facilitate their performance by maintaining consistent quality of the pretreated feed water. Current pretreatment technologies for desalination are designed to reduce the fouling potential of the feed water by removing natural organic matter and suspended solids. However, pretreatment technologies typically are not designed to remove dissolved solids [50].

Inorganic scaling in membrane and thermal desalination processes caused by sparingly-soluble dissolved salts in the feed water limits the operating conditions and the overall system recovery of these processes. In membrane desalination processes, scaling limits the degree to which the feed water may be concentrated (i.e. limits system recovery) [99] and establishes corresponding limits on the system operating pressures and fluxes. In MED and MSF, scaling reduces heat transfer efficiency, restricts operating temperatures, and lowers the system recovery [100–102].

In order to prevent the deleterious effects of scaling, FO can function as an advanced pretreatment technology that removes both dissolved organic material and dissolved inorganic scalants from the feed water, in addition to suspended constituents. When FO pretreatment is used, the coupled conventional desalination process for draw solution recovery is only exposed to an engineered draw solution that has negligible fouling and scaling potential, such as a sodium chloride solution or an ammonia–carbon dioxide solution. As discussed in Section 5.1, the reversibility of fouling in FO indicates that under proper hydrodynamic conditions, FO membranes can maintain their flux and performance when in contact with raw feed waters that have a high fouling potential. A schematic of FO pretreatment for a conventional membrane or thermal desalination process is shown in Fig. 9.
Using FO as a pretreatment can enhance the performance of conventional desalination processes by removing sparingly-soluble scalants from the feed water. Coupled desalination processes may achieve higher system recoveries by operating at higher pressure or temperature without the risk of scaling. Process modeling of an FO–RO system and bench-scale testing of both FO–RO and FO–NF systems have demonstrated the practicality of implementing FO as a pretreatment process. The potential benefits of FO pretreatment for achieving higher system recoveries in membrane desalination were demonstrated by pilot-scale studies of brackish water desalination with ion exchange pretreatment and seawater desalination with NF pretreatment. The advantage of FO pretreatment over ion exchange or NF, in addition to the lower fouling propensity, is the ability to remove all dissolved species from the feed water, not just specific cations or anions.

Simulation of MSF and MED thermal processes for seawater desalination showed that FO pretreatment significantly reduced the concentrations of multivalent ions in the pretreated feed water, thus reducing the scaling tendencies in these processes and enabling them to operate at higher temperatures and recovery rates. Process modeling of FO pretreatment for thermal desalination technologies builds upon extensive modeling and pilot-scale studies of NF pretreatment for these technologies. Modeling results and data from pilot-scale testing of NF pretreatment for MSF showed that the scaling potential was reduced and higher operating temperatures were possible. FO pretreatment rejects all dissolved constituents from the feed water, which is an advantage over NF pretreatment. In addition, NF increases operating costs and fouling propensity because it is a pressurized system.

6.4. FO pretreatment can improve the permeate quality produced by reverse osmosis

The ability of FO pretreatment to remove dissolved constituents from the feed water can also help conventional RO desalination meet stringent product water quality requirements. The degree to which a dissolved solute is rejected by desalination membranes, like those used in FO and RO, is affected by characteristics of the solute, characteristics of the membrane, and complex interactions between the feed water and membrane surface. Two types of dissolved contaminants for which incomplete rejection by RO is a concern for potable water production and water reuse are trace organic compounds (TrOCs) and boron. TrOCs in the aquatic environment are derived from human sources, and they have been the subject of regulatory interest and public concern. Boron exists naturally in seawater as uncharged boric acid, and its removal is important to protect human health and to enable eventual reuse of the desalinated seawater for agricultural irrigation. FO pretreatment provides an additional barrier for dissolved constituents, reducing the concentrations of TrOCs and boron in the product water compared to conventional RO desalination.

FO demonstrated higher rejection of representative TrOCs than RO in bench-scale studies, and hybrid FO–RO systems have demonstrated excellent rejection of TrOCs (>99%) because of the dual membrane barrier to these contaminants. FO has also been demonstrated to more effectively reject boron than RO because of the influence of reverse solute flux in FO. Current practice is to employ additional RO passes in series to achieve high boron rejection in seawater desalination. However, process modeling of FO pretreatment for seawater RO desalination indicates that boron removal could be achieved more energy-efficiently with an FO–RO system than a conventional two-pass RO process. While the FO–RO system shows promise for TrOC and boron removal, different rejection rates for TrOCs or boron in the FO and RO processes may result in their accumulation in the draw solution of a closed-loop FO–RO system and may potentially degrade product water quality. Adsorption by activated carbon, ultraviolet light oxidation, and draw solution bleeding have been suggested as mechanisms to mitigate contaminant accumulation in the closed-loop draw solution.

6.5. Osmotic dilution applications of FO are truly low energy

Osmotic dilution is the concept of utilizing the salinity difference between two solutions to drive water permeation in FO, without a draw solution recovery process. In osmotic dilution, a dilute stream...
becomes concentrated and a concentrated stream is diluted as permeation occurs across a semipermeable FO membrane. Both the diluted and concentrated solutions are the products of osmotic dilution, and no draw solution recovery is necessary. By eliminating the energy-intensive draw solution recovery, osmotic dilution is truly a low-energy FO process and can achieve the “free lunch” referenced in Section 2.1.

Osmotic dilution is often conceived as the coupling of an impaired feed water (e.g., wastewater effluent) with saline sources (e.g., brackish or seawater) to concentrate the impaired feed water and dilute the saline feed. Osmotic dilution can be potentially adapted at two stages in a conventional seawater desalination facility to (i) dilute the feed water to the RO process to a lower salinity, resulting in reduced energy requirement, and (ii) dilute the concentrated brine prior to discharge to mitigate detrimental impacts to the environment [23]. Osmotic dilution offers the potential for energy and cost savings in an RO facility by lowering the operating hydraulic pressure. Environmental impacts may be diminished by reducing electricity requirements, and also by discharging brines with lower salinity to the aquatic ecosystem. Moreover, reducing the volume of the impaired water offers an additional benefit [24].

Although osmotic dilution has been demonstrated to be an effective process at the bench-scale [23,74] and pilot-scale [23,76], osmotic dilution continues to face challenges before it is commercially implemented. An osmotic dilution system requires a large membrane area due to the relatively low osmotic pressure difference and resulting low water flux, which corresponds to large footprint and capital costs [121]. Additionally, the use of an impaired water for osmotic dilution of the feed water to a drinking-water treatment facility, such as a seawater reverse osmosis facility, continues to be perceived negatively by consumers [24], limiting deployment of this practice.

7. Concluding remarks

Through simple thermodynamic arguments, we have shown that FO cannot reduce the minimum energy required for desalination, regardless of the type of draw solution used. An FO-RO desalination system cannot consume less energy than a standalone RO system to achieve a certain recovery. However, FO hybrid systems can outperform other desalination technologies when they are applied to the desalination of high-salinity feed waters using thermolytically driven solutions to consume less total energy for separation.

Research into FO membrane fabrication has yielded commercially-available, high water flux FO membranes with low structural parameters. While extensive research on FO continues to be conducted with commercial asymmetric CTA membranes, successful applications of FO will utilize TFC polyamide membranes specifically engineered for FO that demonstrate relatively high permeability and selectivity. High selectivity is crucially important for FO membranes for operational, economic, and environmental reasons. In particular, FO treatment of high-salinity feed waters requires high osmosic pressure draw solutions generated by low molecular weight draw solutes, further underscoring the need for FO membranes with very high selectivity. Simple dissolved inorganic and thermolytic salts remain the most effective draw solutes because they generate high osmotic pressures to drive permeation and maintain high diffusivities to mitigate ICP. The design of highly selective TFC membranes is constrained by the inherent permeability-selectivity tradeoff, and we expect that the best-performing TFC-FO membrane will have modest water permeability, high selectivity, inert surface chemistry, and an ultra-low structural parameter.

One apparent advantage of the FO process is the relatively lower fouling propensity and higher fouling reversibility compared to RO. A thermodynamic energy analysis of the FO-RO process does not capture this important benefit of FO, which can potentially outweigh the energetic costs of using FO in hybrid systems and may enable these systems to treat feed waters with high fouling potential. Discerning fouling mechanisms in FO and developing fouling mitigation strategies will allow FO to reach its full commercial potential in a variety of applications. Development of FO membranes should continue to be focused on increasing fouling resistance. FO excels with challenging feed waters, and its future applications will be in treating high-salinity or high fouling potential waters, or waters that contain specific contaminants like boron or TrOCs. The unique advantages of FO that will drive these future applications are its low fouling propensity, capacity for high osmotic pressure driving forces that exceed the operating limits of RO, and high rejection of feed water contaminants. Hybrid FO systems employing thermolytic draw solutions, such as the ammonia–carbon dioxide system, can be used to desalinate high-salinity feed waters while consuming less total energy than applicable thermal desalination technologies. FO may also be applied as advanced pretreatment to improve water quality and enable higher system recovery for conventional desalination technologies by rejecting feed water foultants, scalants, and contaminants of concern.

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