



Recent developments and future challenges of forward osmosis for desalination: a review

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ABSTRACT

This article focuses on the recent developments of forward osmosis (FO) for desalination and the challenges it faced. Issues discussed include membrane, configuration, draw solution, integration with post-treatment, and energy and economic evaluation. In spite of the potential advantages of FO, there exist a number of technical barriers that impede FO's application for water desalination. Further membrane development is needed to give high water flux, low reverse solute flux and material stability over the operating pH range. It is currently not economically viable to use NaCl as the draw solute for municipal scale FO desalination in view of the solute loss through the membrane. Progress in post-treatment is also needed, because FO cannot stand as a single process, but has to be integrated with a post treatment to regenerate draw solution and produce desalinated water that meets the required standards. A FO–RO process is technically feasible, but economically unviable due to energy consideration. In order for FO to compete with the existing RO desalination technology, breakthrough is necessary in the three key areas of membrane development, selection of appropriate draw solution, and integration with a suitable post-treatment, and these need to be addressed based on a total system approach.

Keywords: Forward osmosis; Desalination; Draw solution; Post-treatment; Energy consumption; Product quality

1. Introduction

Osmosis, or forward osmosis (FO) process is a physical phenomenon known as the net transport of water across a semi-permeable membrane driven by a difference in chemical potential across the membrane, i.e., passage of water is from the lower to higher solute concentration side. Although the FO phenomenon was observed 260 y ago [1], little studies were conducted before the progress of membrane technology especially

reverse osmosis (RO) was made in 1960's. The interests in research on FO have increased rapidly in the past ten years. Fig.1. Shows a research publication trend of FO, based on searching results from Engineering Village.

The increasing attention on FO is mainly due to its potential to achieve high level of rejection for a wide range of contaminants without the need to apply hydraulic pressure for separation [2,3]. As such, FO may compare favorably to pressure-driven processes such as RO in that it may have lower demand on electrical energy, and potentially less foulant compaction since no hydraulic pressure is applied. Not surprisingly, FO

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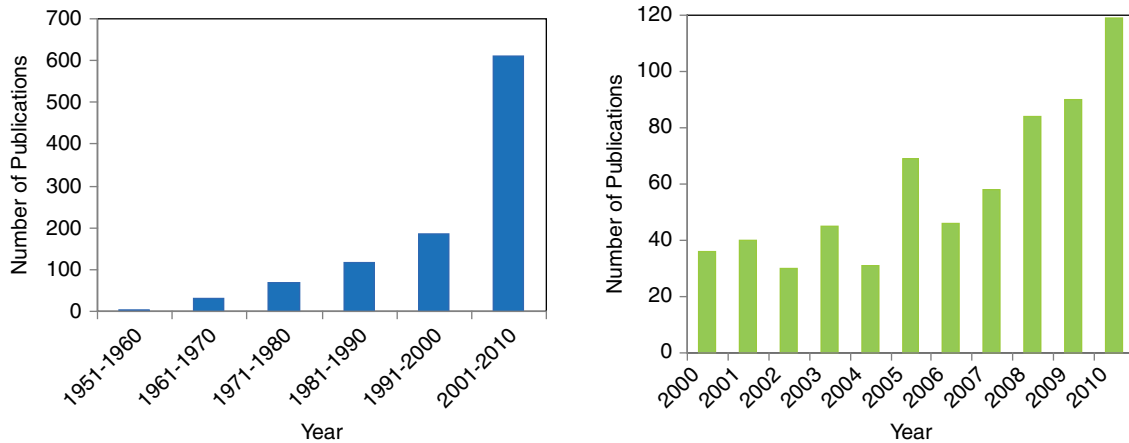


Fig. 1. Papers published on FO.

has been reported to have potential for a wide range of applications encompassing treatment and reclamation of wastewater, seawater/brackish water desalination, pressure-retarded osmosis for generation of electricity, food processing and controlled drug release as well as for RO backwash cleaning [2–5]. The focus of this review is to provide the state-of-the-art of the physical principles and recent developments of FO for seawater desalination, as well as future challenges that it faces in application.

2. Principles of forward osmosis for seawater desalination

In a FO process, water passes through a semi-permeable membrane from lower to higher salt concentration side while solute molecules or ions are rejected. The driven force is actually a difference in chemical potential because the lower the concentration, the higher the water chemical potential. In practice, the osmotic pressure difference between both sides of the FO membrane is used to describe the driving force for water transport although it does not need to apply a physical/hydraulic pressure on the membrane during FO process. However, the net driving force in a RO process is subtraction of the osmotic pressure difference ($\Delta\pi$) from the hydraulic pressure applied (ΔP). Fig. 2. illustrates the difference in driving force between FO and RO.

The theoretical osmotic pressure π of a solution can be derived using the Van't Hoff Equation as follows:

$$\pi = n \cdot c \cdot \phi \cdot R \cdot T \quad (1)$$

where, n is the number of ions; c is salt concentration (M), ϕ is osmotic coefficient; R is the universal gas constant (0.082057 l·atm/K/mol), and T is temperature (K).

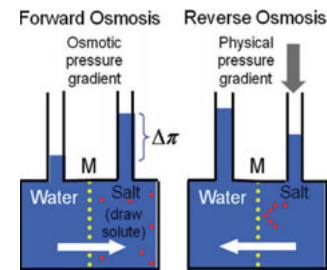


Fig. 2. Driving force in FO and RO processes.

Without the effect of hydraulic pressure, the water passage through the FO membrane can be generally described by Eq. 2:

$$J_w = A \cdot \sigma \cdot \Delta\pi \quad (2)$$

where J_w is the water flux ($l/m^2/h$); A is the water permeability coefficient ($l/m^2/h/atm$) of the membrane which is a measure of how easily water transports across the FO membrane; σ is the reflection coefficient; $\Delta\pi$ is the effective osmotic pressure gradient (atm) cross the membrane between the draw and feed solution sides which is the driving force of FO [3].

In addition, concentration polarization (CP) is an important issue in FO process. It is believed that the coupled effect of dilutive internal concentration polarization (DICP) and concentrative external concentration polarization (CECP) limits FO membrane flux [3,5–7] as shown in Fig. 3. CECP can be negligible when the feed flows in a turbulent pattern, but DICP can be a limiting issue [5,7]. DICP occurs within the porous support layer of the membrane due to dilution of the draw solution ($C_{D,b} \rightarrow C_{D,i}$) caused by the water flux permeated from the active layer, resulting in reduction of the effective

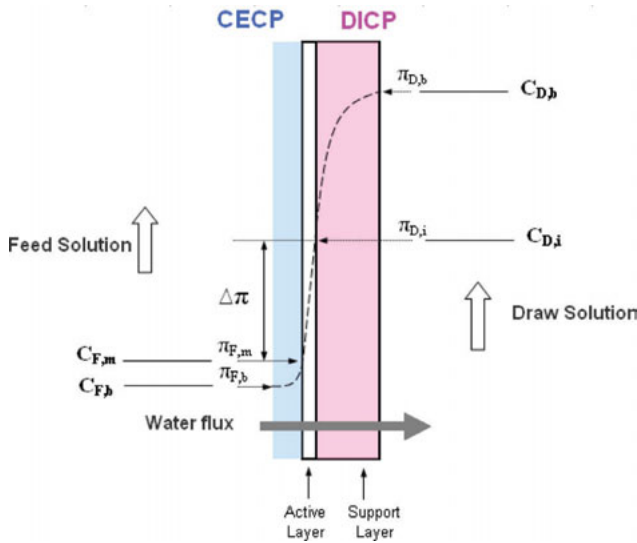


Fig. 3. Illustration of the coupled DICP and CECP. Adopted from [7] with permission.

osmotic pressure ($\pi_{D,b} \rightarrow \pi_{D,i}$) as the driving force. The effective osmotic pressure at the draw solution side can be expressed as [5]:

$$\pi_{D,i} = \exp(-J_w \cdot K) \cdot \pi_{D,b} \quad (3)$$

where, K is the solute resistivity (d/m) for diffusion within the porous support layer and is defined as [3]:

$$K = \frac{t\tau}{D\varepsilon} \quad (4)$$

where, t is thickness of the support layer of the membrane, τ is tortuosity of the support layer, ε is porosity of the support layer, and D is solute diffusion coefficient (m^2/s).

Therefore, to achieve a large effective osmotic pressure driving force, the effect of DICP must be reduced by increasing $\pi_{D,i}$ or minimizing K value. This means that the substrate or support layer of the FO membrane is to be thin (small t) and of high porosity (large ε), and at the same time, the draw solute is to have a high diffusion coefficient (large D), so that the K value and hence DICP can be minimized [3,7]. From Eq. (2), the water flux of FO can also be enhanced by increasing A , which requires a high performance FO membrane with a thin dense skin of high salt rejection and low water permeation resistance.

Theoretically, as per Eq. 2, the water flux may be increased by utilizing a strong draw solution with higher osmotic pressure. However, a larger water flux will also intensify DICP as predicted by Eq. 3, which acts

to reduce the effective driving force. As a result, increasing draw solution concentration will only have a diminishing effect on flux enhancement, as the relationship between flux and osmotic pressure of the draw solution is logarithmic [3,8].

The reverse diffusion of draw solute or salt—as typically the case—is a naturally occurring phenomenon that accompanies a FO process, and needs to be minimized. This reverse draw solute diffusion happens because solute concentration in the draw solution is much higher than that in the feed solution. The diffusion of solutes (J_s) through a FO membrane can be described by Fick’s Law in Eq.5:

$$J_s = B \cdot \Delta c \quad (5)$$

Here, B is the solute permeability coefficient and Δc is the solute concentration difference across the membrane. The reverse draw solute diffusion depends on draw solution composition and FO membrane characteristics. The reverse draw solute diffusion not only reduces the driving force but also contaminates the feed solution. Depending on the material properties of the draw solute of concern, the reverse draw solute diffusion may have an impact on the treatment and disposal of the concentrate stream.

From Eqs. (5) and (2), the ratio of the reverse solute flux (J_s) to the water flux (J_w) can be derived to give the following relationship for the assumption that σ is approximately 1 [9]:

$$\frac{J_s}{J_w} = \frac{B}{A} \cdot \frac{1}{n \cdot \phi \cdot R \cdot T} \quad (6)$$

This relationship demonstrates that the J_s/J_w ratio is directly dependent on the membrane separation characteristics (B/A) for a given operating condition ($n\phi RT \sim \text{constant}$). More on discussion on membrane development with the desired separation characteristics will be provided in Section 3.1, the J_s/J_w ratio also has a direct consequence on the operating costs of a FO plant as it determines the amount of draw solute that needs replacement (m_s) [10]. For a given product flow rate (Q_p), the following relationship can be derived:

$$m_s = \frac{J_s}{J_w} \cdot Q_p \quad (7)$$

The above discussion makes plain that reverse draw solute diffusion into the feed solution is a critical area that needs further research for FO application.

Fig. 4 shows a schematic of a FO desalination process. As illustrated, FO cannot be a standalone process

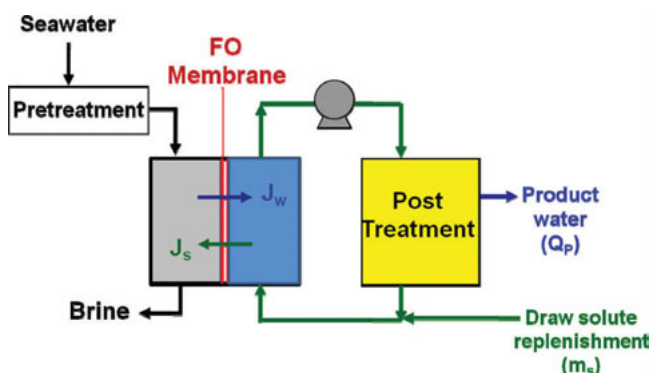


Fig. 4. Schematic of FO desalination process.

by itself for the purpose of desalination. Minimally, it has to be integrated with a post-treatment step to be a functioning desalination system, and for practical application, a pretreatment step may also be necessary. How a FO desalination process functions may be described as follows. After the necessary pre-treatment, the FO step will extract pure water out of the seawater through the semi-permeable membrane to the draw solution side, while a wide range of contaminants are retained in the brine stream. Driving force for this water transport is the osmotic pressure gradient between the seawater (in which water has higher chemical potential) and the more concentrated draw solution (in which water has lower chemical potential). In a subsequent post-treatment step, the draw solution that is mixed with the permeated water will be treated to give the clean product water and to simultaneously regenerate the draw solute for the FO step. Any shortfall in the draw solute will need to be replenished accordingly in the draw solution circuit.

In spite of the prevailing interest in FO, there are a number of key challenges that impede FO's application for municipal scale water desalination to compete with the existing RO technology. The first key challenge is the need for development of high performance FO membrane that is capable of maintaining high water flux and low reverse solute diffusion under real operating conditions. Another key challenge is the selection of appropriate draw solution that needs to be of significantly higher osmotic pressure than seawater (around 25 atm for 3.5% salt concentration) and at the same time meets other criteria for application such as low cost and safe to handle. Finally, the post-treatment step needs to be able to cost-effectively separate water from the draw solute and to fully recycle the draw solute by minimizing energy consumption. In order for FO to be competitive with the existing RO technology, the overall energy requirement of the FO system may not exceed 4 kWh/m³, which is the baseline for seawater RO operations. Note that these

challenges are inter-related in that membrane optimization needs to be considered against the type of draw solute used, and this, in turn, has to be considered against the adopted post-treatment step, and vice-versa.

Therefore, further development of the FO technology for seawater desalination will need to address the three above-mentioned aspects: (1) FO membrane; (2) draw solute (also known as osmotic agent); and (3) integration with compatible post-treatment, and this has to be accomplished based on a total system approach due to their inter-relationships.

3. Developments of forward osmosis for seawater desalination

3.1. Membranes for forward osmosis

Although the osmosis phenomenon was observed by Nollet in 1748 [1], no progress on membrane development was made. Early studies focused on the mechanism of osmosis through natural materials. Special attention has been given to FO only with the development of synthetic membrane materials since the first Loeb–Sourirajan asymmetric cellulose acetate RO membrane with high flux and high salt rejection was developed in 1960's [11].

In general, any dense, non-porous, and selectively permeable material can be used as a membrane for FO. Such flat sheet and hollow fiber membranes [5,12–28] have been tried for various applications of FO in the past forty years.

Batchelder [13] was the pioneer using natural cellulose as a membrane material for FO trial in 1965. Frank in 1972 [14] explored seawater desalination with cellulose acetate (CA) RO membrane. Votta et al. in 1974 [15] and Anderson in 1977 [16] tested several commercially available and an in-house CA RO membranes to treat dilute wastewater by FO using a simulated seawater draw solution. Kravath and Davis in 1975 [17] utilized CA flat sheet RO membranes from Eastman and hollow fiber membranes from Dow to desalinate seawater by FO with glucose as the draw solution. Goosens and Van-Haute in 1978 [18] used CA RO membranes reinforced with mineral fillers to evaluate whether membrane performance under RO conditions can be predicted through FO testing. However, performance of the membranes tested mentioned above were not explored. Mehta and Loeb in 1979 [19] tested the FO performance of flat sheet DuPont B-9 and hollow fiber B-10 Permasep RO membranes made of an aromatic polyamide polymer. There was no FO membrane available in the 1970s. Only RO membranes (either flat sheet or hollow fiber configuration) were used in all investigations on the FO process and much lower flux was observed than theoretical values predicted from the classical solution diffusion

theory in all cases [3]. The low flux was attributed to the fact that RO membranes typically consist of a very thin active layer but a thick porous support layer (a few hundred micro meter) which caused a very large internal CP [12].

In the 1990s, a special FO membrane with significant improvement in water flux was developed by Osmotek Inc. (now Hydration Technology Innovations LLC (HTI)) [29]. Research on FO has been increasingly attractive since then. SEM images of this type of FO membrane are shown in Fig. 5. This proprietary membrane is made of cellulose triacetate (CTA). The thickness of the membrane is less than 50 μm and it is obvious that the structure of the CTA-FO membrane is quite different from a standard RO membrane. A unique feature of the CTA-FO membrane is its lack of a thick support layer. Instead, the embedded polyester mesh provides mechanical support for the membrane. This type of CTA-FO membrane has been used successfully in commercial applications of water purification for military, emergency relief and recreational purposes [29,30]. The membranes have also been extensively investigated for seawater desalination with ammonia carbon-dioxide as osmotic agents [31,32], which will be introduced in detail in Section 3.3. In addition, they have been tested

in a wide variety of applications by different research groups [33–38]. However, the CTA-FO membrane material is not expected to tolerate low or high pH. Martinetti et al. [39] confirmed that Na_2EDTA solution at pH 11.8 did degrade the CTA membranes during cleaning.

Various studies had demonstrated that the CTA-FO membranes supplied by HTI performed significantly better with water flux of an order of magnitude greater than conventional RO membranes when used for FO application. When normalized to the osmotic pressure, CTA-FO membranes could typically achieve osmotic pressure-corrected flux in the range of 0.1–0.5 $\text{l/m}^2/\text{h}/\text{atm}$, as compared to values around 0.01 $\text{l/m}^2/\text{h}/\text{atm}$ for typical RO membranes [5,12,19–27,32]. The higher water fluxes achieved by the CTA-FO membranes could be attributed to its relative thinness without a fabric support layer, which helped in mitigating internal CP. The reverse solute flux reported for the CTA-FO membranes ranged 0.6–11 $\text{g/m}^2/\text{h}$ for NaCl as the draw solute. There were differences in the reported water flux and reverse solute flux because different types or batches of CTA-FO membranes were tested under different operating conditions [27]. In general, the reverse solute flux increases with an increase in the water flux as predicted by Eq (6).

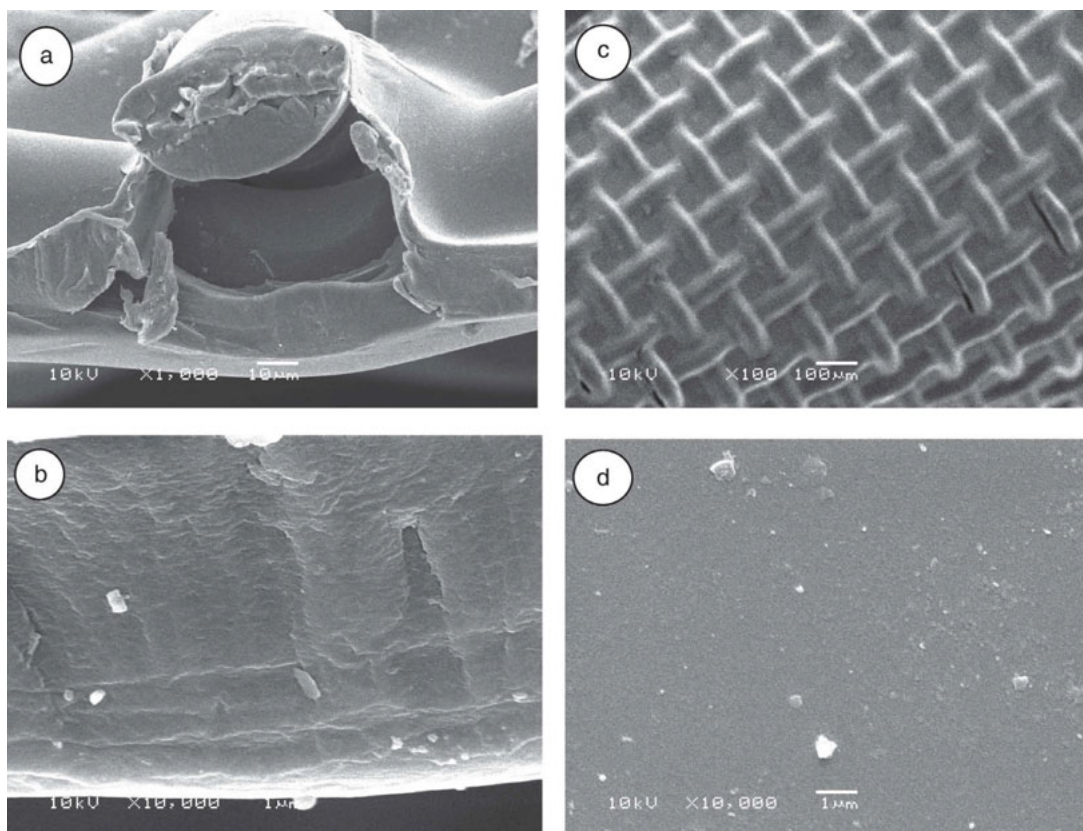


Fig. 5. SEM images of HTI's FO membrane (a. Cross-section: 1000x; b. Top skin:10,000x; c. Back: 100x; d. Skin surface: 10,000x).

Currently, most FO membranes used are in flat sheet configuration. Since two cross-flow channels are required at both sides of the FO membrane, hollow fiber configuration is more suitable for FO desalination process, as one may simultaneously induce/force flow on both sides of a hollow fiber membrane in a simpler manner [40–43]. In addition, hollow fiber membranes have the advantages of self-support and high packing density compared to flat sheet configuration. Therefore, development of high performance FO hollow fiber membranes is a priority area for further research for FO desalination.

An overview of a few selected recent examples of membrane development for FO is given in Table 1.

Table 1 shows that three main groups of FO membranes have been developed. These are the CTA-FO membrane, polybenzimidazole-based (PBI) FO membrane and thin-film composite (TFC) FO membrane.

Main attributes of the CTA-FO membrane had already been discussed in the foregoing paragraphs [23,27,32]. Additionally, it may be noted from [10] that despite having different draw solution concentrations, J_s/J_w and A are generally constant, as these are related to the intrinsic properties of the membrane. The fact that $J_w/\Delta\pi$ reduces with increasing draw solution concentration for the same CTA-FO B membrane is due to the effect of internal CP as revealed in Eq (3).

Recently, in Chung's research group, a new type of PBI hollow fiber nanofiltration membrane has been developed for FO process [22,42,43]. PBI has robust mechanical strength and excellent chemical stability compared to CTA-FO membranes. The PBI-based membranes have undergone improvement [42]. Currently, a novel FO hollow fiber membrane with PBI-PES dual-layer structure has been fabricated using a co-extrusion technology with promising result and significantly improved water permeability [43]. While the PBI-based membranes generally exhibited low rejection values to monovalent ions (NaCl), they demonstrated high rejection values to divalent ionic compounds ($MgCl_2$), which could be attributed to ion size exclusion and Donnan electrostatic effect [22,42,43]. The PBI-based membrane developments are currently still in the laboratory scale.

Wang et al. [28] have successfully developed two types of TFC FO membranes with an ultra-thin RO-like skin layer on either the inner surface or outer surface of a porous PES hollow fiber substrate by a two-step preparation in the laboratory. The formation of a RO-like skin layer on the outer or inner surface of the PES substrate was made based on interfacial polymerization [44]. The TFC-FO hollow fiber membranes gave higher water flux and lower reverse solute flux compared to the CTA-FO flat sheet membrane (see Table 1).

Other developments include Catalyx Inc (Anaheim, California) [45], which claimed that the Catalyx

Table 1
Overview of recent FO membrane developments

Year	Membrane	Draw solution	Water flux J_w [l/m ² /h]	Reverse solute flux J_s [g/m ² /h]	Solute flux: water flux ratio J_s/J_w [g/l]	Osmotic pressure corrected water flux [#] $J_w/\Delta\pi$ [l/m ² /h/atm]	Water permeability coefficient [#] A [l/m ² /h/atm]	Ref.
2005	AG-RO	6M NH_4HCO_3	≤ 2	–	–	≤ 0.01	Similar A value	[31]
	CE-RO		≤ 2			≤ 0.01		
	CTA-FO		23			0.1		
2008	CTA-FO	0.5M NaCl	8.5	7.4	0.9	0.4	–	[23]
2009	CTA-FO A	50 g/l NaCl	15	17	1.1	0.4	–	[27]
	CTA-FO B		11	8	0.7	0.3		
	CTA-FO C		6	1	0.2	0.2		
2009	PBI-based dual layer NF	5M $MgCl_2$	33.8	0.55	0.02	0.06	1.74	[43]
2010	TFC-FO A	0.5M NaCl	12.9	2.3	0.2	0.6	0.95	[28]
	TFC-FO B		32.2	3.7	0.1	1.4	2.22	
2010	CTA-FO B	50.8 g/l NaCl	12.2	9.1	0.75	0.3	0.78	[10]
		35.2 g/l NaCl	9.6	7.2	0.74	0.4		
		17.9 g/l NaCl	6.2	4.6	0.74	0.5		

[#]Although the osmotic pressure-corrected water flux ($J_w/\Delta\pi$) and the water permeability coefficient (A) have the same units in l/m²/h/atm, their derivation and meaning differ. The $J_w/\Delta\pi$ shows how the driving force is being utilized for flux production, whereas A measures intrinsic water transport property of a membrane under hydraulic pressure in RO mode.

FO membrane would be able to minimize internal CP phenomenon by having a high porosity, yet self-supporting membrane in an extremely thin layer. It was reported that Catalyx had developed a hybrid FO–RO system with NaCl as a draw solution for wastewater treatment but not for seawater desalination. Specifications of the membrane however were not reported. In addition, Yip et al. [46] developed high performance thin-film composite FO membranes that showed water fluxes over 18 l/m²/h with 1.5 M NaCl as draw solution and pure water as feed, and achieved salt rejection more than 97%. The membrane performance remained stable after prolonged exposure to an ammonium bicarbonate draw solution. The possibility of developing biomimetic membrane for FO application is another opportunity that merits attention and further support [47].

In summary, the desired characteristics of FO membranes are very thin and high dense skin layer for high water flux (high A) and high solute rejection (low B); a thin substrate with maximum porosity for minimal internal CP (small K); hydrophilicity of the skin layer for reduction of membrane fouling; and tolerance to chemicals for cleaning and draw solution. Currently, only Hydration Technologies Inc. provides the commercially available CTA-FO membranes. However, the CTA-FO membranes only have a narrow range of pH tolerance for cleaning, whereas other types of FO membranes are still at the stage of laboratory development. Further breakthrough in the development of improved FO membranes is critical for advancing the technology for seawater desalination.

3.2. Draw solutions (osmotic agents) for FO

As mentioned in Section 2, osmotic pressure gradient between the draw solution and the feed provides the driving force for a FO process. Therefore, selection of appropriate draw solution or osmotic agent is critical to achieve high FO water flux. Furthermore, an ideal draw solution needs to fulfill the requirements [2,3,10]: (a) high solubility in water and relatively low molecular weight, so as to achieve high osmotic pressure efficiency; (b) being inert, stable, neutral or near neutral pH, nontoxic; and (c) being easily and inexpensively separated to yield potable water and to be recycled; and (d) low-cost and compatible to the membrane with low reverse solute flux. In particular, the last requirement is crucial for the viability of the process at the municipal scale, and has direct consequence on the operating costs of a FO plant.

Fig. 6. Shows the daily amount of draw solute that needs to be replenished for a FO process due to loss through the membrane for three different J_s/J_w scenarios. The figure clarifies why the commonly used NaCl may

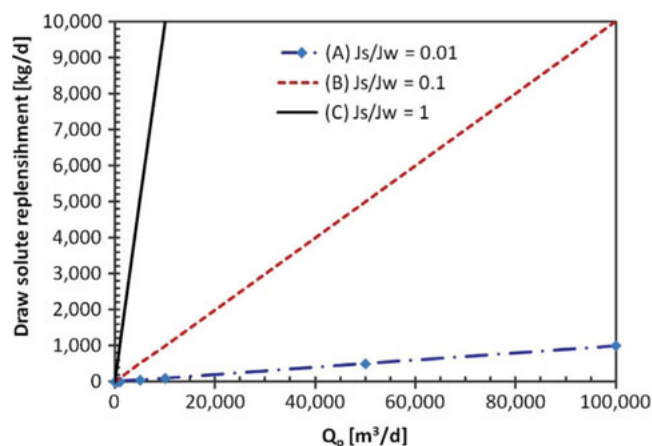


Fig. 6. Daily requirement of draw solute replenishment [kg/d] as function of water production [m³/d] derived based on Eq. (7) for the three scenarios: (A) $J_s/J_w = 0.01$ g/l; (B) $J_s/J_w = 0.1$ g/l; and (C) $J_s/J_w = 1$ g/l.

not be adopted for municipal scale FO application at this point of time. Current FO membranes could achieve J_s/J_w down to around 0.1 g/l for NaCl (Scenario B in Figure). This means that a municipal scale FO plant with the capacity of 100,000 m³/d will lose 10,000 kg of NaCl on a per day basis, which need replenishment. From a logistics point of view, Scenario C needs no further consideration for a municipal scale FO plant, whereas Scenario A is technically not yet achievable with the current FO membranes and for NaCl as the draw solute.

Another way of evaluation is to consider the additional operating costs to a FO desalination plant due to replenishment for the lost draw solutes for scenarios of various draw solute cost and J_s/J_w ratio (Table 2). The table presents a 3×3 matrix according to the draw solute price (rows) and J_s/J_w ratios (columns). Typically, costs for various types of draw solutes range between \$10 per kg and \$100 per kg [10], such that commonly available chemicals such as NaCl may be represented by the \$10 per kg cost range (Row II in Table), whereas other more specialized chemicals may be represented by the \$100 per kg cost range (Row III in Table). It is also assumed that specific low cost draw solute at the \$1 per kg cost range (Row I in Table) will be developed in the future.

Range of desalination cost for a municipal scale plant (size > 60,000 m³/d) is typically 0.5 – 1 \$ per m³ of water produced [48]. On the basis that the additional operating costs due to replenishment for lost draw solutes should not exceed 10 – 20% of the desalination cost for a municipal scale FO plant, a practical cost limit for draw solute replenishment may be set as 0.1 \$/m³. From Table 2, it is evident that only the matrix entries I-A, I-B, and II-A would fulfill this criterion (as bolded in the table). Commonly available chemicals such as NaCl may land in

Table 2
Additional operating costs to a FO desalination plant for replenishment of lost draw solutes

Draw solute cost	J_s/J_w Ratio		
	(A) $J_s/J_w = 0.01$ g/l	(B) $J_s/J_w = 0.1$ g/l	(C) $J_s/J_w = 1$ g/l
(I) 1 \$/kg	0.01 \$/m³	0.1 \$/m³	1 \$/m ³
(II) 10 \$/kg	0.1 \$/m³	1 \$/m ³	10 \$/m ³
(III) 100 \$/kg	1 \$/m ³	10 \$/m ³	100 \$/m ³

the matrix entry II-B, which corroborates the above analysis on why it is currently not viable to use NaCl as the draw solute for municipal scale FO desalination. In the same way, economical viability for a municipal scale FO desalination plant may not be achieved with an expensive draw solute with very low J_s/J_w (III-A in the table); neither with a low-cost draw solute with high J_s/J_w (I-C

in the table). The discussion here makes clear that both J_s/J_w and draw solute cost are crucial factors for selecting an appropriate draw solution for a FO application.

Different types of draw solute, also known as osmotic agent, have been studied in the past four decades. An overview of draw solutes with different recovery/separation methods is tabulated in Table 3.

Table 3
Overview of draw solutes with different recovery methods studied in FO

Year	Draw solute (osmotic agent)	Recovery method	Ref.
1965	SO ₂	Heating or air stripping	13
1965	Alcohols, SO ₂	Distillation	49
1972	Al ₂ SO ₄	Doped Ca(OH) ₂ to precipitate CaSO ₄ and Al(OH) ₃	14
1975	Glucose	None	17
1976	Concentrated solution of nutrients	None	50
1989	Fructose	None	51
1992	Glucose/sucrose	Low pressure RO	52
1997	MgCl ₂	None	10, 12
2002	KNO ₃ and SO ₂	Precipitate KNO ₃ by cooling. SO ₂ is removed through standard means	53
2002	NH ₃ & CO ₂	Heating	54
2005	NaCl	Osmotic distillation	20
2005~ 2006	NH ₄ HCO ₃	Heating NH ₄ HCO ₃ , decompose into NH ₃ & CO ₂	5, 31, 32, 55
2006	NH ₄ HCO ₃	None	56
2007	Magnetic nanoparticles	Separated by using a canister separator	57
2007	Albumin	Denatured and solidified	57
2007	Dendrimers	Using a wide range of pH values for recovery	57
2007	Magnetic nanoparticles	Magnetic field	58
2008	MgSO ₄ , NaCl, NaNO ₃ and ZnSO ₄	None	10, 23
2008	Cross-linked poly(acrylic acid) as superabsorbent	Hydraulic pressure	59
2009	NH ₄ HCO ₃	None	10, 62
2010	NaHCO ₃ , KHCO ₃ , Na ₂ SO ₄	RO	10
2010	2-Methylimidazole based solutes	Membrane distillation (MD)	60
2011	Fertilizers	None	61

Batchelder [13] was the pioneer to patent using volatile solutes (sulfur dioxide) as a draw solution to extract water from seawater in 1965. The volatile solutes are removed by a heated gas stripping operation. The patent, however, only determined that positive water flux occurred in the experiments and did not quantify the flux or salt rejection. Glew in 1965 [49] used an approach similar to that idea. In the patent, he utilized the mixtures of water and sulfur dioxide or liquid (e.g., aliphatic alcohols) as the draw solution for FO. The gas and liquid were added in the water in order to reduce the activity of the water. The osmotic agents were then removed from the fresh water by distillation. That was the first FO technique that attempted to remove and recycle the draw solute in the overall process.

Frank in 1972 [14] proposed another draw solution consisting of precipitate-able soluble salt (aluminum sulfate) as osmotic agent. The salt could be treated using calcium hydroxide to separate water from the precipitates (calcium sulphate and aluminum hydroxide). The excess calcium hydroxide in the water could be neutralized via addition of sulfuric acid or carbon dioxide.

Kravath in 1975 [27], Kessler in 1976 [50] and Stache in 1989 [51] explored a concentrated solution of nutrients such as glucose and/or fructose as osmotic agents to extract pure water from seawater. The osmotic agents were not recycled and this would result in a less concentrated drinkable nutrient solution, but not fresh water. Their objective was to develop a batch desalination process for emergency water supply on lifeboats, not as a continuous process for seawater desalination at a municipal scale. Yaeli in 1992 [52] employed a mixture of glucose and sucrose as an osmotic agent for FO desalination. The large size of the sucrose made it possible to be recovered by RO.

From late 1990's, more studies have been started to investigate salts as osmotic agents. Loeb in 1997 [12] tested $MgCl_2$ to conduct a fundamental study of FO process without recovery of the solute. McGinnis in 2002 [53] utilized the temperature-dependent solubility of potassium nitrate and sulfur dioxide as draw solutions with a two step process for seawater desalination. Later, McGinnis [54] disclosed that combination of ammonia and carbon dioxide gases in a specific ratio could produce a draw solution with high osmotic pressure in excess of 250 atm. The new approach could remarkably increase water flux and water recovery. Based on this idea, Elimelech and co-workers have intensively investigated ammonium bicarbonate (NH_4HCO_3) as a draw solute for seawater desalination since 2005 [5,31,32,55]. The low molecular weight and high solubility of NH_4HCO_3 could bring about higher water fluxes. Furthermore, the draw solute could be decomposed to ammonia and carbon dioxide gases at 65°C by heating and be recovered.

Ng et al. [56] also conducted a fundamental study of FO with NH_4HCO_3 draw solution. Cornelissen et al. in 2008 [23] reported a lab FO study with $MgSO_4$, NaCl, $NaNO_3$ and $ZnSO_4$ as draw solutes.

In addition, magnetic nanoparticles, such as magnetoferritin surrounded by polyethylene glycol [57] and hollow protein spheres [58], had been tested as osmotic agents in FO, as the magnetic nanoparticles are water soluble and can provide reasonable osmotic pressure. The nanoparticles could be recovered from the purified water using a magnetic separator. However, development of suitable magnetic nanoparticles with high osmotic pressure is a challenge. Adham et al. also reported albumin and dendrimers as candidate draw solutions with UF post-treatment in a lab study [57]. However, the osmotic pressure of albumin is too low to provide adequate driving force for seawater desalination and both compounds are very expensive.

Recently, Jones et al. in 2008 [59] patented a cross-linked superabsorbent polymer as an osmotic agent. The superabsorbent polymer, such as poly(acrylic acid), poly(methacrylic acid) and their neutralized salts, is water swell-able but not water soluble. The absorbed water could be released when a mechanical pressure would be exerted on the surface of the polymer. Recently, Chung et al. [60] tried 2-Methylimidazole based organic solutes as osmotic agent and post-treated the draw solution by membrane distillation (MD). This study demonstrated the potential application of a draw solute that undergoes thermal decomposition at temperature of 315°C in an integrated FO-MD process in the laboratory. However, the recovered draw solute showed a significant increase in reverse solute flux (2.4 g/m²/h) compared to that of the fresh draw solute (0.9 g/m²/h). The FO process achieved a relatively low flux of <6 l/m²/h with a 5M draw solution, and a synthetic feed seawater with 3.5% NaCl.

Achilli et al. have evaluated 14 types of draw solution for FO application [10]. They found that $CaCl_2$, $KHCO_3$, $MgCl_2$, $MgSO_4$ and $NaHCO_3$ could be very good draw agents from a performance analysis, while $KHCO_3$, $MgSO_4$, NaCl, $NaHCO_3$, and Na_2SO_4 fared favourably from a replenishment cost analysis. Relatively, $KHCO_3$, $MgSO_4$ and $NaHCO_3$ may have the potential to fulfil both criteria of high performance and low replenishment cost.

Hancock et al. [26] investigated solute-coupled diffusion in FO process using CTA-FO membranes and NH_4HCO_3 as draw solution in a laboratory study. They found that the reverse solute flux of NH_4HCO_3 was far greater than that of NaCl. Based on the reported J_s/J_w ratio of 2.9 g(NH_4HCO_3)/l (water) [26], estimation of the solute (salt) leakage loss through the CTA-FO membrane would be 290 tons/d for a municipal scale plant with

the capacity of 100,000 m³/d, and would pose a challenge for practical application as can be inferred from Fig. 6.

More recently, a novel low energy fertilizer driven FO desalination has been developed for the irrigation application. However, there are limitations with the concept [61]. In such a process, fertilizers are used as draw agents for FO process because the diluted draw solution can be directly applied for fertilization. This concept provides several novelties: (1) the cost of desalinated water is very low; (2) an additional separation/recovery process for draw solutions is not necessary; and (3) the process directly provides nutrient-rich water for irrigation of crops and plants. However, the fertilizer draw solution could only extract water from a saline solution up to the concentration where the osmotic potentials of both feed and draw solution are equal. Furthermore, the final fertilizer concentration needs to be reduced to the acceptable limit for using water from other sources.

In spite of efforts in the past four decades, further developments in draw solution remain a critical factor necessary to advance the FO technology. Up till now, no ideal draw solution has yet been invented to satisfy all criteria to make it viable to apply FO for seawater desalination. In particular, an appropriate draw solution needs to possess high osmotic pressure efficiency and low reverse solute flux. In this regard, draw solution selection and membrane development have an interdependent relationship in order to achieve a desirably low J_s/J_w ratio.

3.3. Integration of FO with post-treatment for desalination

Numerous trials have been attempted to desalinate water based on the FO technique in the past four decades, however, few progresses have been made due to the challenges faced as described in Sections 3.1 and 3.2. Several patents have also been awarded for different methods of integrating FO with post-treatment as a total system for desalination [13,14,51–54,58,59]; however, most of these have not been proven feasible. The followings provide a discussion based on selected examples.

Cath et al. [20] investigated the energy consumption with a pilot-scale hybrid FO-RO system for reclamation and reuse of wastewater for space application, where CTA-FO membranes were used in the FO pretreatment prior to RO. The RO configuration was 4 pass, and serves as post-treatment to FO for producing potable water required in space missions. The feeds were synthetic wastewater stimulants including hygiene wastewater, humidity condensate and urine and the draw solution was NaCl solution. The energy consumption was measured during operation under variable operating conditions. The specific energy consumption ranged

from 15 to 30 kWh per m³ of purified water. The high power consumption was mainly due to the 4 pass RO configuration applied.

The research group at Yale University has also intensively investigated applying FO process for seawater desalination in the laboratory [3,5,21,31,32,55]. In the process, commercial available CTA-FO membranes were used. Ammonium bicarbonate (NH₄HCO₃) was utilized as the osmotic agent for the FO process, and a thermal process such as distillation was proposed as the post-treatment to strip and recover the osmotic agent. The energy requirements of ammonia-carbon dioxide FO desalination were predicted by the use of chemical process modeling software (HYSYS) [62]. The FO process was modeled using single or multiple distillation columns to separate draw solutes from the product water for recycling within the FO system. Thermal and electrical energy requirements of the process were provided. It was reported that the electrical power requirement of FO alone in seawater desalination would be typically less than 0.25 kWh/m³, however, the heat requirement with the single distillation would be more than 270 MJ/m³ (or >75 kWh/m³). It is also well understood that the energy requirement for temperature increase by 1°C is 1.16 kWh for every m³ of water at 100% conversion efficiency. It is therefore evident that such an integrated FO process would make practical sense, only if adequate supply of waste heat is available. In January 2007, it was reported that Yale University would construct the FO desalination pilot plant with completion expected by Spring of 2007 [63]. However, data of the pilot testing with real seawater feed have not yet been reported at the moment.

Oasys Water Inc. [64] reported that Oasys, formed with a seed investment from GreatPoint Ventures, was to further develop its FO desalination technology, known as Engineered Osmosis™ (EO). The EO employs the natural osmotic pressure gradient created by a highly concentrated ammonia/carbon dioxide 'draw solution' to pull water from a feed water stream through a membrane. The ammonia and carbon dioxide could then be easily separated from product water using low-grade waste heat from sources such as power generation plants. The startup has so far established a pilot-scale plant to test the technology with a production capacity of 1 m³/d. However, it is not clear whether the pilot tests were conducted with synthetic feed water or real seawater. The product quality was also not reported. While pilot tests were ongoing, it was announced that a demonstration plant would be completed in the fourth quarter of 2010.

It was reported in 2007 [65] that a Manipulated Osmosis Technology developed by University of Surrey that leverages on a two-stage FO-RO desalination

process could potentially reduce energy consumption to <1.6 kWh/m³ with proprietary osmotic agents used as draw solution for FO. The proprietary process had been demonstrated using a 1 m³/h pilot unit and would be now ready for commercialization. However, the report was not clear whether the feed of the desalination process was seawater or brackish water, which would have significant impact on the process efficiency and energy consumption due to difference in the osmotic pressure and hence the available driving force. More detailed information about this technology is also not publicly available at the moment. In general, usage of a “strong” draw solution with high osmotic pressure, on one hand, creates a high driving force and results in a high water flux (productivity) in FO. On the other hand, the “strong” draw solution will significantly increase the energy consumption of the RO post-treatment because the diluted draw solution would have a much higher osmotic pressure than the seawater feed (about 24 atm). In other words, it is a challenge for a hybrid FO–RO process to be viable for desalination and to compete with conventional RO technology in terms of energy consumption.

Khaydarov from INP developed a solar powered direct osmosis desalination process in 2007 [66]. Diethyl ether (C₂H₅)₂O with low boiling point of 35.6°C was used as the osmotic agent and evaporation with a heat exchanger was applied for recovery of the osmotic agent. A pilot plant of 1 m³/h capacity was tested at site with brackish water feed of 17 g/l. It was reported that the specific electric energy consumption of the desalination process was less than 1 kWh/m³ with solar energy consumption of 80 MJ/m³ (or 22 kWh/m³). The total dissolved solid (TDS) in the product stream after treatment was about 50 mg/l. However, the residual diethyl ether in the product was not reported. It was also observed that the membrane used had relatively short lifetime (not more than 2–3 mo), because the polymeric membrane material could be destroyed by the organic osmotic agent (diethyl ether).

In 2009, QuantumSphere [67] developed a FO process using a certain organic solution as the draw solution to separate water from salt water. The process uses a semi-permeable membrane to separate water from salt water into the special organic solution across the membrane. The diluted organic solution is then heated to allow the specially formulated organic solute to be removed, leaving only fresh drinking water after a final purification step by activated charcoal. It was claimed that the FO process so developed could potentially purify water at less than 3000 kWh per acre ft (or 2.43 kWh/m³). It was also reported that the company had constructed a prototype system with the capacity of several gallons/d for demonstration purposes and intended to scale up the process

with large development partners to serve large-scale municipal drinking supplies for seawater desalination. However, the report was not clear, whether the organic solute could be fully recovered for recycling and what would be the energy consumption required for separation of the organic from water by heating.

Choi et al. [68] investigated a FO and RO hybrid system for seawater desalination with a laboratory-scale device. A simple film theory model was applied to consider CP in FO and RO processes for the estimation of internal and external CP effects in FO process. The modeled flux was compared to experimental flux under a variety of operating conditions. It was found that the combination of FO and RO may result in a higher flux than FO-only process under some operating conditions. However, the energy efficiency of FO and RO hybrid system was not studied.

Another novel hybrid FO–RO process for drinking water augmentation using impaired water and saline water sources was also developed [69]. In the process, FO was used to extract water from an impaired aqueous feed solution while seawater was applied as the draw solution. By using the hybrid process, the seawater would become diluted to be fed to SWRO, resulting in a lower energy consumption of desalination, while leveraging on the two barriers of both FO and RO membranes to prevent contaminants present in the impaired water from migrating into the product water. FO was also studied as an option to treat RO brine streams [70]. It was found that water recovery of up to 90% in FO could be achieved from the brine streams and high water flux of FO could be maintained effectively for an extended time by adding a scale inhibitor. However, another RO was again necessary for post-treatment of the FO permeate to produce fresh product water and simultaneously to regenerate the draw solution (52g/l NaCl) for the FO. The issue of concern here is the high energy consumption that would be expected. Similar studies on integrated FO processes for desalination were also reported [71–75].

Danasamy [76] carried out a laboratory study on applying FO for seawater desalination using CTA-FO membranes and ammonium bicarbonate (NH₄HCO₃) solution (2.5 M) as the draw solution. Unique of this study is that real seawater with TDS of 30g/l was used as the feed, compared to other studies that used synthetic saline water as feed [5,31,32,55,56,62]. It was surprisingly observed that the TOC in the FO permeate was nearly 10 mg/l. Further analysis indicated that the TOC was non volatile organic and it was due to the NH₄HCO₃ draw solution (2.5 M). It was also found that the boron removal in FO was 40–60% and boron concentration in the FO permeate was >1 mg/l. Furthermore, it was observed that FO water flux with real seawater as the

feed was 3 times lower than that with synthetic seawater (0.5 M NaCl) as the feed while all other experimental conditions remained the same. This study demonstrated that membrane fouling would be a serious issue for practical FO application, which so far had not been discussed extensively in the literature. Another issue of concern is the final product water quality. For NH_4HCO_3 -type draw solutions, the residual ammonium concentration in the product water needs further study.

To date, no integrated FO system has shown to be sufficiently viable for seawater desalination. Further advancement of the technology will also require the development of a suitable post-treatment, and this has to be optimized under consideration of the applied draw solution as a total system. In order to be viable, an integrated FO desalination system needs to have overall energy consumption lower than 4 kWh/m^3 , which is the baseline for existing RO desalination technology.

4. Conclusions

Fundamental research on FO and the development of new applications of FO have been steadily growing in the recent decades, because FO may have advantages of very low hydraulic pressure operation, high rejection of a wide range of contaminants, and potentially low membrane fouling tendency. However, there exist a number of technical barriers that impede FO's application for water desalination because FO cannot stand as a single process alone for a municipal scale application. In order for FO to compete with the existing RO desalination technology, breakthrough is necessary in the three key areas of membrane development, selection of appropriate draw solution, and integration with a suitable post-treatment, so as to achieve an overall FO system that gives low energy consumption and good product quality.

In terms of membrane development, FO membranes need to provide high water permeation, high rejection of solutes, minimal internal CP, high chemical stability, and high mechanical strength. There is still a lack of robust FO membranes and modules that are adequate for desalination. Currently, only CTA-FO membranes are commercially available, but these membranes are not able to tolerate a wide range of pH under operating conditions. Promising FO membranes are currently in development, but these are still in the laboratory stage and not yet ready for application. In terms of membrane configuration, the trend is hollow fiber, which is hydrodynamically compatible with water flowing on both sides of the membrane, and it allows for high packing density. The development of improved membrane and module is crucial for advancing the FO technology for water desalination.

Draw solute, or osmotic agent, for FO application needs to meet the criteria: high osmotic efficiency, stable, near neutral pH, nontoxic, membrane compatible, easily separable, low solute leakage, and inexpensive. For desalination, a key challenge is to be able to derive a draw solution that possesses greater osmotic pressure than the seawater itself (around 25 atm for 3.5% salt concentration), and concomitantly, is cost-effective in terms of replenishment and recovery. Selection of an appropriate draw solution for FO application is a critical area that needs more research and requires optimization in conjunction with membrane development and post-treatment integration.

Integrated FO systems with different post-treatment methods for water desalination have been studied. A few showed potential, but most of these systems are not feasible for municipal scale application due to issues concerning low process efficiency, high energy consumption and product water quality that does not meet standards. In summary, FO is currently not yet a viable option for municipal scale desalination. To advance the technology, critical limitations concerning membrane development, draw solution and cost-effective post-treatment need to be overcome, and these need to be addressed based on a total system approach.

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