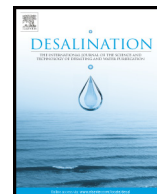




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Effect of reverse solute diffusion on scaling in forward osmosis: A new control strategy by tailoring draw solution chemistry

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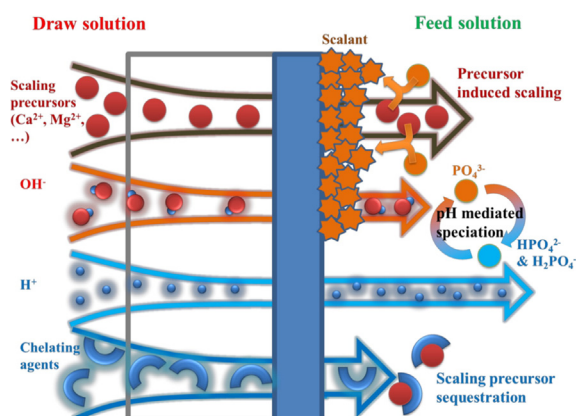
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HIGHLIGHTS

- Reverse solute diffusion (RSD) has important effect on FO scaling.
- RSD of scaling precursors (Ca^{2+} , PO_4^{3-} , etc.) can promote severe $\text{Ca}_3(\text{PO}_4)_2$ scaling.
- RSD of anti-scaling agents (H^+ , EDTA, etc.) can suppress $\text{Ca}_3(\text{PO}_4)_2$ scaling.
- The effect of RSD of solute i is negligible if $(J_s/J_v)_i \ll (C_{feed})_i$.

GRAPHICAL ABSTRACT



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ABSTRACT

We explored the specific role of reverse solute diffusion (RSD) on the scaling in osmotically-driven membrane processes (particularly forward osmosis (FO)). Both scaling precursors (e.g., Ca^{2+} and phosphate) and anti-scaling precursors (e.g., H^+ and a chelating agent ethylenediamine tetraacetic acid (EDTA)) were used to investigate the effect of RSD and draw solution chemistry on calcium phosphate scaling. While draw solutions containing Ca^{2+} tend to promote calcium phosphate scaling, this effect was noticeable only if the specific RSD of Ca^{2+} (i.e., the ratio of Ca^{2+} flux to water flux) was greater than the original Ca^{2+} concentration in the feed water. The RSD of H^+ and EDTA effectively suppressed scaling. For the first time, we demonstrated a new scaling control strategy for FO by the inclusion of anti-scaling functions in the draw solution chemistry. Our work has important implications for the design and operation of FO processes.

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1. Introduction

Osmotically driven membrane processes (ODMPs), including forward osmosis (FO) and pressure retarded osmosis (PRO), have gained more attention in the fields of seawater and brackish water desalination

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[1–3], wastewater reclamation [4,5], resource recovery [6,7], liquid food processing [8], and electricity generation [9,10]. Like reverse osmosis (RO) and nanofiltration (NF), FO may suffer from the scaling of sparingly soluble inorganic minerals [11], thus causing reduced water flux and shortened life span of membranes [12–14] or even the loss of membrane integrity [15,16]. Some common membrane scalants include calcium phosphate in wastewater reclamation and calcium carbonate and calcium sulphate in desalination [17,18].

According to the existing literatures, scaling of RO and NF membranes is affected by feed water chemistry (saturation index (SI), pH, and ionic strength), operating conditions (permeation flux, applied pressure, crossflow velocity, and temperature), and the presence of other competing agents such as antiscalants and organics with chelating properties [14,19–21]. Similar behaviors have been reported for FO scaling [22–26]. However, scaling in ODMPs can be further complicated by internal concentration polarization (ICP) and reverse solute diffusion (RSD). Zhang et al. [27] reported that PRO scaling in active-layer facing draw solution orientation (AL-DS) was greatly enhanced by (1) the RSD of scaling precursors from the DS to the feed solution (FS) and (2) the concentrative ICP of both draw solutes (through RSD) and feed solutes inside the porous membrane substrate. Although concentrative ICP is not important in the active-layer facing feed solution orientation (AL-FS, which is commonly used for FO applications), this orientation will still suffer from the RSD effect [11].

Both inorganic and organic DSs have been reported in the literature [28,29]. According to some recent reviews [29–31], the major requirements for an ideal DS include high osmotic pressure, high water solubility, low viscosity, high diffusivity, and easy regeneration. Nevertheless, its propensity in promoting membrane fouling should not be overlooked [27,32,33]. It has been reported that some specific ions (e.g., SO_4^{2-} and Ca^{2+}) in DS can act as precursors to promote severe membrane scaling during PRO operation [27]. However, the exact conditions under which this effect is important has not been systematically investigated. In addition, the water chemistry of a DS can also potentially affect the pH of the feed solution (e.g., via the RSD of H^+ or OH^-), which can either reduce or promote scaling. Conceptually, by carefully tailoring DS chemistry and imparting anti-scaling/antifouling functionalities to the DS, one may be able to control scaling (as well as other types of fouling) in ODMPs effectively – an option that is never available for pressure-driven membrane processes. Such considerations may open a new dimension for DS design and selection. To the authors' best knowledge, the use of DS chemistry for reducing FO scaling has not been reported previously.

The objective of this paper is to systematically investigate the specific role of RSD on FO scaling. Calcium phosphate, a commonly found sparingly soluble mineral in wastewater, was used as model scalant due to its significance in membrane-based wastewater reclamation [24,34,35]. The role of specific ions in DS and the inclusion of anti-scaling chemistry were systematically tested. The results of the current study may provide important insights into FO scaling and scaling control.

2. Material and methods

2.1. Chemicals and solutions

Unless specified otherwise, all solutions were prepared with ACS grade chemicals and MilliQ water. FS and DS were prepared by the addition of pre-determined amounts of NaCl, CaCl_2 , Na_3PO_4 , $\text{NH}_4\text{H}_2\text{PO}_4$, $(\text{NH}_4)_2\text{HPO}_4$, and Ethylenediaminetetraacetic acid disodium salt (EDTA). Their solution pHs were adjusted using NaOH and HCl. In the current study, the EDTA was chosen as a model chelating agent to sequester calcium from participating in scaling formation. The DS compositions were chosen in such a way as to achieve an identical initial FO water flux ($17.5 \pm 0.8 \text{ L/m}^2 \cdot \text{h}$). Detailed compositions and properties of the FSs and DSs are summarized in Tables 1 and 2, respectively. OLI's

Table 1
Compositions and properties of feed solutions.

Feed solution	NaCl (mM)	CaCl_2 (mM)	Na_3PO_4 (mM)	pH	Osmotic pressure (bar) ^a
Baseline tests	61	0	0	7.5	2.8
Scaling tests w/o Ca^{2+} in FS	35	0	20	7.5	2.8
Scaling tests with Ca^{2+} in FS	30	12	0.8	7.5	2.8

^a All the feed solutions have identical osmotic pressure. The osmotic pressure is calculated using the OLI's Stream Analyzer 3.1 software (OLI Systems, Inc., Morris Plains, NJ).

Stream Analyzer 3.1 (OLI System, Inc., Morris Plains, NJ) was used for the calculation of osmotic pressure values.

2.2. Membranes

Two commercial FO membranes (a cellulose triacetate membrane denoted as CTA and a thin film composite polyamide membrane denoted as TFC) used in the study were obtained from Hydration Technologies Inc. (HTI, Albany, OR). The TFC membrane had a higher water permeability and solute rejection (Table 3) but also a greater surface roughness [36–38]. In comparison, the CTA membrane had a much smoother surface [37]. It had been well documented in the literature that a rougher membrane surface tends to promote fouling [11,39].

2.3. Bench-scale FO scaling tests

A crossflow FO setup was modified from our existing study [40]. Briefly, an FO membrane coupon with an effective area of 42 cm^2 was placed in a CF042 (Sterlitech) cell. Diamond-patterned net-type spacers were inserted into both the FS and DS channels. A crossflow velocity of $\sim 9.7 \text{ cm/s}$ was used for both the FS and DS. The temperature of both feed water and draw solution were at ambient temperature ($25 \pm 1 \text{ }^\circ\text{C}$). The weight change of DS was measured by a digital balance, which was used for FO water flux calculation.

All scaling tests were performed in the AL-FS orientation. A new piece of membrane was used for each scaling test. After the 20-min conditioning of the membrane coupon by a calcium-free feed solution in test cell, a desired amount of CaCl_2 was introduced into the FS to initiate the scaling test. Where needed, some minor pH adjustment was also performed immediately after the dosage of CaCl_2 to ensure the solution pH was maintained at the desired value. The scaling test was then continued for another 8 h and membrane flux was measured by monitoring the weight change of the feed solution using a digital balance. Baseline tests were also performed using feed solutions of identical osmotic pressure (using NaCl solution) and pH (Table 1). The deviation of water flux between the scaling tests from the corresponding scaling-free baseline level can be attributed to FO scaling [41]. To study the effect of chemistry of different draw solutions, their compositions were chosen in such a way to ensure an identical initial membrane flux (see detailed DS composition in Table 2). Scaled membranes were further characterized using a Joel JSM 7600F Field Emission Scanning Electron Microscope (SEM). Air-dried membrane coupons were sputter-coated with a thin

Table 2
Compositions and properties of draw solutions.

Draw solution	Concentration	pH	Osmotic pressure (bar) ^a
NaCl	2.0 M	4.0, 7.0, or 8.5	100
CaCl_2	1.3 M	7.0	116
$\text{NH}_4\text{H}_2\text{PO}_4$ + NaCl	1.0 M + 1.0 M	4.5	92
$(\text{NH}_4)_2\text{HPO}_4$ + NaCl	1.0 M + 1.0 M	8.5	94
EDTA + NaCl	0.1% + 1.0 M	7.0	100

^a The draw solutions were chosen in such a way that an identical initial FO flux of $17.5 \pm 0.8 \text{ L/m}^2 \cdot \text{h}$ was obtained for all tests. The osmotic pressure is calculated using the OLI's Stream Analyzer 3.1 software (OLI Systems, Inc., Morris Plains, NJ).

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