Effect of hydraulic pressure and membrane orientation on water flux and reverse solute flux in pressure assisted osmosis

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A B S T R A C T

Forward osmosis (FO) is an emerging technology that has received much global interest due to its potential applications in wastewater reclamation and seawater desalination. One of the major challenges facing the FO process is the detrimental effects of concentration polarization (CP), which reduce the effective osmotic pressure driving force and thus decrease productivity of the FO process. In this study, pressure assisted osmosis (PAO) was investigated as a method to increase the effective driving force and water flux by combining an osmotic pressure driving force with an additional hydraulic pressure. Experiments were carried out to examine the efficiency of the PAO process using a bench-scale setup specially designed to prevent membrane deformation under the applied hydraulic pressure. Results showed that PAO water flux increased with increasing the applied hydraulic pressure in FO mode (i.e., active layer facing the feed solution). The measured water fluxes were in good agreement with predictions based on a model developed to describe the water flux in PAO operation. However, the PAO water flux was lower than model predictions in PRO mode (i.e., active layer facing the draw solution). This observation is attributed to the spacer ‘shadow effect’ and the resulting reduction in the effective membrane area by the spacers. The results also showed that reverse solute flux decreased with increasing the applied hydraulic pressure in both FO and PRO modes. Although applying hydraulic pressure to FO increases energy consumption, the higher water flux in PAO reduces the number of membrane modules for the FO process. In addition, control of the driving force is easier in PAO than FO, leading to flexibility in system design and operation. Based on these results, a possible combination of FO and RO system with PAO was proposed for allowing higher energy efficiency in seawater desalination.

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1. Introduction

Increasing concern over the limited supply of fresh water from conventional sources has increased the need to develop alternative water sources, including the reuse of wastewater and seawater desalination. Forward osmosis (FO) is an emerging membrane technology that can be used to produce clean water from these alternative water sources. FO is driven by the osmotic pressure difference across a semi-permeable membrane, and has received much interest in recent years because of the wide range of potential applications [1–3]. Compared with pressure-driven membrane processes, FO may offer several advantages, including lower use of electric energy, high rejection of contaminants, and low membrane fouling tendency [4]. Examples of potential FO applications include wastewater treatment [5–7], seawater/brackish desalination [8,9], and energy generation using salinity gradients [10–16]. However, there are several limitations that prevent large-scale applications of the FO process. One critical limitation is the phenomenon of concentration polarization (CP), which reduces the effective osmotic pressure driving force across the membrane, thereby lowering water flux. Of particular importance is the internal concentration polarization (ICP) within the support layer of FO membranes, which results in a marked decrease in the osmotic pressure driving force compared to external concentration polarization (ECP). Due to ICP, the FO water flux and productivity
are generally lower than those of pressure-driven membrane technologies, such as reverse osmosis (RO). Because CP is inherent of the FO process and cannot be eliminated, it is of paramount importance to better understand the phenomenon and to develop methods to reduce its deleterious effects on the FO process.

In this paper we investigate a new FO operation, pressure assisted osmosis (PAO), which incorporates the FO and RO processes. In PAO, in addition to the osmotic pressure driving force originating from the draw solution, hydraulic pressure is also applied to increase the driving force for water transport. The additional hydraulic pressure on the feed side was shown to not only increase permeate water flux, but also reduce reverse solute flux. We focused on the performance of PAO, with an emphasis on the effect of hydraulic pressure and membrane orientation. Based on the results obtained, we proposed a possible process innovation incorporating PAO for seawater desalination.

2. Modeling water flux

Following the general equation for FO presented by Lee et al. [13], McCutcheon and Elimelech [17] derived the following simplified equations to predict the water flux, \(J_w\), in FO in the presence of ECP and ICP:

\[
J_w = A \left( \frac{\pi_{D,b} \exp \left( -\frac{J_w}{K} \right)}{\pi_{F,b} \exp \left( \frac{J_w}{K} \right)} \right) \quad \text{(for FO mode)} \tag{1}
\]

\[
J_w = A \left( \frac{\pi_{D,b} \exp \left( -\frac{J_w}{K} \right)}{\pi_{F,b} \exp \left( \frac{J_w}{K} \right)} \right) \quad \text{(for PRO mode)} \tag{2}
\]

\[
K = \frac{\tau}{D} = \frac{S}{D} \tag{3}
\]

Here, \(A\) is the membrane pure water permeability, \(\pi_{D,b}\) and \(\pi_{F,b}\) are the draw and feed solution bulk osmotic pressures, respectively, \(K\) is the solute resistance to diffusion within the porous support layer, \(k\) is the mass transfer coefficient in the boundary layer at the active layer side of the membrane, \(t\), \(r\), and \(\epsilon\) are the thickness, tortuosity, and porosity of the support layer, respectively, \(D\) is the bulk diffusion coefficient of the draw solute, and \(S\) is the structural parameter of the membrane. The mass transfer coefficient \(k\) is obtained from

\[
k = \frac{ShD}{d_h} \tag{4}
\]

where \(Sh\) is the Sherwood number and \(d_h\) is the membrane channel hydraulic diameter. The Sherwood number for a rectangular membrane channel can be obtained as described elsewhere [16].

In Eqs. (1) and (2), the solute resistivity, \(K\), and mass transfer coefficient, \(k\), are the key factors that control the permeate water flux, \(J_w\). Choi et al. proposed new equation for combined system of FO and RO as below [18].

\[
J_w = A \left( \frac{\Delta P + \pi_{D,b} \exp \left( -\frac{J_w}{k_D} \right)}{\pi_{F,b} \exp \left( \frac{J_w}{k_D} \right)} \right) \quad \text{(for FO mode)} \tag{5}
\]

where \(\Delta P\) is the transmembrane applied hydraulic pressure, and \(k_D\) and \(k_{D,0}\) are the mass transfer coefficients for ECP and ICP, respectively. Note that in this equation, \(k_D\) and \(k_{D,0}\) are equivalent to \(k\) and \(1/K\) in Eqs. (1) and (2).

It is noteworthy that the total water flux in PAO is not simply the sum of the water fluxes by FO and RO (i.e., \(J_w = J_{w,FO} + J_{w,RO}\)). Actually, the increased water flux by the applied hydraulic pressure decreases effective osmotic pressure due to enhanced ICP as predicted by the \(\pi_{D,b} \exp \left( -\frac{J_w}{k_D} \right)\) term in Eq. (5).

For our experiments with deionized (DI) water feed in FO and PRO modes, the equations for water flux simplify to

\[
J_w = A \left( \Delta P + \pi_{D,b} \exp \left( -\frac{J_w}{k_D} \right) \right) \quad \text{(for FO mode)} \tag{6}
\]

\[
J_w = A \left( \Delta P + \pi_{D,b} \exp \left( -\frac{J_w}{k_{D,0}} \right) \right) \quad \text{(for PRO mode)} \tag{7}
\]

Here, we neglected the osmotic pressure term of feed side because DI water was used as feed solution (\(\pi_{F,b} = 0\)). Further, the concentration of salt in feed solution caused by reverse solute flux was extremely small compared to draw solution concentration, even at the end of the experiment; hence, this assumption (i.e., \(\pi_{F,b} = 0\)) is acceptable in our experiments.

3. Material and methods

3.1. Membrane

Commercially available FO membranes (HTI, Albany, OR) made of cellulose triacetate (CTA) were used in this study. They are flat sheet membranes with an asymmetric structure. Polyester woven mesh is embedded in the membrane structure to improve mechanical strength. Basic information on this membrane is given elsewhere [19].

3.1.1. Membrane characterization

The \(A\) and \(B\) coefficients were measured using an identical membrane sample. The two coefficients were measured every time when a membrane was replaced by a new sample after each run.

3.1.2. Membrane orientation

Generally, two membrane orientation modes exist in the FO process: one with the active layer facing the feed solution (FO mode) and the active layer facing the draw solution (PRO mode). Membrane orientation is important because water flux behaviors are different for different membrane orientations, even with identical concentrations of draw solution (i.e., identical osmotic pressure) [20]. In our experiments, when the applied pressure was over 20.7 bar (300 psi) in PRO mode, the membrane was damaged. On the other hand, membrane damage was not observed in FO mode, even at 27.6 bar (400 psi). For this reason, the maximum applied pressures were set to 13.8 bar (200 psi) in PRO mode and 20.7 bar (300 psi) in FO mode, respectively, in our experiments.

3.2. Spacer and new test cell

Fig. 1 shows the schematic of the specially fabricated cell and spacer design used in our experiment. Spacers are composed of two different materials. One is a porous stainless steel spacer, 1.75 mm in thickness, that allows water to pass freely. The width and length of the spacer are designed to fit the channel dimensions in the test cell. The other spacer is a tailored permeate carrier, which is used in commercially available spiral wound RO membrane modules (Hydraulantics Inc.). Four sheets of permeate carrier are used to fill the remaining channel. These two types of spacers are placed on both the feed and draw solution channels of the test cell.

The spacers play a very important role in our experiments. In PRO experiments, for example, when general diamond shape plastic mesh spacers were used, the membrane underwent severe deformation by the applied hydraulic pressure on the feed side. This membrane deformation phenomenon has been reported recently [11,16]. In our experiments, the same phenomenon was observed and salt rejection decreased remarkably because the
conventional test cells have solution inlet and outlet in the new type of inlet and outlet in the described above, stable membrane was fractured at high pressure. After replacing the stainless steel spacer and a tailored permeate carrier typically used in spiral wound RO modules. In addition to spacers, small holes are drilled on the channel bottom surface in order to minimize empty (unsupported) space on the channel bottom surface.

Fig. 1. Custom-designed cell and spacers for PAO. To support the applied hydraulic pressure on the feed side, two different spacer materials are used: a porous stainless steel spacer and a tailored permeate carrier typically used in spiral wound RO modules. In addition to spacers, small holes are drilled on the channel bottom surface in order to minimize empty (unsupported) space on the channel bottom surface.

membrane was fractured at high pressure. After replacing the diamond shape plastic mesh spacer with the new spacers described above, stable A and B permeability coefficients and salt rejection were observed. The new spacers not only minimized membrane deformation but also helped the membrane to maintain its intrinsic properties.

The test cell is a frame designed with rectangular flow channels on each side of the membrane. The dimensions of the flow channels are 8.1 cm in length, 2.6 cm in width, and 0.3 cm in depth, providing an effective membrane area of 21.06 cm². Conventional test cells have solution inlet and outlet in the flow channel which are too large to support the spacer and membrane against the applied hydraulic pressure [16]. Hence, we designed new type of inlet and outlet in the flow channel. In the new test cell, the solution inlet and outlet are composed of small drilled holes on the channel bottom surface in order to minimize the empty space on the channel bottom surface (Fig. 1). With these specially designed inlet and outlet, we were able to prevent spacer and membrane deformation near the inlet and outlet.

3.3. Experimental setup

Fig. 2 describes the lab-scale cross flow PAO system used in this study. Variable speed gear pump (Cole-Parmer, Vernon Hills, IL) was used to recirculate the draw solution while high pressure pump was used for the feed solution recirculation. The applied hydraulic pressure was controlled by a back pressure regulator placed downstream on feed side. Back pressure on the draw side was monitored with a mechanical pressure gauge while back pressure of the feed side was monitored with a digital pressure gauge.

Concentrations of the draw solution were 0.5, 1.0, and 2.0 M using NaCl (Guarantee reagent, Junsei, Japan) while DI water (SHUVQ-90ST, Ultra-pure water equipment, Sinhan Science Tech, Korea) was used as the feed solution. Salt concentration and temperature change in the feed and draw solution channels were measured by a conductivity meter (HQ40d, Hach Co., Germany). Temperatures of both draw and feed solutions were maintained at 20 ± 1 °C using chillers (AD-RC08, AND, Korea). Flow meters (F-450, Blue-white, USA) were used to measure the volumetric flow rate. The volumetric flow rates in both the feed and draw channels were set 1 l/min at the initial stage, but the flow rate changed on the draw solution side after hydraulic pressure was applied due to the reduction in the flow channel effective height of the membrane test cell as described later. Permeate flux was measured every 3 min using micro scale (Cuw 4200H, Cas, Korea) which was connected to a computer for automatic data logging.

3.4. PAO run

Before PAO tests, the A and B coefficients of the membrane were measured first. Initially, the membrane was compacted at 13.8 bar using DI water. After flux was stable (usually no more than 30 min), the permeate flux was measured in FO mode at the following applied pressures: 6.9, 13.8, 17.25, and 20.7 bar (100, 200, 250, and 300 psi). For tests in PRO mode, we employed the following applied pressures: 3.45, 6.9, 10.35, and 13.8 bar (50, 100, 150, and 200 psi). The pure water permeability coefficient A was calculated from the slope of water versus pressure plot.

We have calculated the reverse solute flux using the following equation [21,22]:

\[
c_{T}(V_{p0} - J_{w} A_{m} t) = J_{s} A_{m} t
\]  

where \( c_{T} \) is the NaCl concentration in the feed, \( V_{p0} \) is the initial volume of feed solution, \( J_{w} \) is the measured water flux, \( A_{m} \) is the membrane area, and \( t \) is time. The PAO performance tests were conducted after evaluating the A and B coefficients using three different salt (NaCl) concentrations and two membrane orientations. For the PAO experiments, we have used the same membrane coupon as we confirmed that repeated experiments do not affect the intrinsic properties (A and B) of the membrane. PAO tests started under no hydraulic pressure (FO system condition) and then the applied pressure increased stepwise (6.9, 13.8, 17.25, and 20.7 bar) at 30 min intervals in FO mode. Similarly, for the PRO mode, the applied pressure increased stepwise (3.45, 6.9, 10.35, and 13.8 bar) at 30 min intervals. Since spacer-filled channels hinder the volumetric flow rate, pressure drops occurred on both the draw and feed channels (about 1.7 bar for FO system condition and about 2.1 bar for PAO system condition). These pressure drops were considered when applying hydraulic pressure in PAO.

4. Results and discussion

4.1. Flux behavior of PAO in FO mode

As indicated earlier, one may simply assume that the water flux in PAO is the sum of two independent different flux components: an osmotic flux and a pressure-driven flux. However, PAO flux is not as high as one may expect because the additional hydraulic pressure also increases the dilutive internal concentration polarization. As a result of increased ICP, the effective osmotic pressure
is reduced and thus the PAO water flux is less than the sum of the osmotic flux and the pressure-driven flux. Accordingly, Eq. (6) should be used to predict the water flux in PAO.

The PAO test results for experiments carried out in FO mode are shown in Fig. 3. All experimental conditions were identical, except for the draw solution concentration as indicated in the figure. The modeled water flux was calculated using Eq. (6) using the average solute resistivity, K. Initial water flux and K values obtained from FO mode experiments under non-pressure conditions are summarized in Table 1. As shown in Table 1, K values varied slightly at each concentration of draw solution, so the average K value was used for modeling. Overall, as seen in Fig. 3, our results show relatively good agreement between measured and simulated water flux values.

Using the water flux data in Fig. 3, the K and S values were calculated as shown in Table 2. Note that S values are different for different concentrations of draw solution. In principle, the membrane structural parameter S should be identical when it is measured using an identical membrane, although it can vary with measurement conditions or method used within a narrow range [17]. However, based on our results, it is evident that the S value decreased as the draw solution concentration increased. Compared with the effect of draw solution concentration, the applied hydraulic pressure does not seem to affect the S value significantly. The S values in our study ranged from 468 to 615 μm, which is in the range of S values reported in the literature. One possible explanation for the different values is the dependence of the diffusion coefficient on salt concentration. We also note that variations of S values are common [16], and such variations were reported in the literature even when using the same membrane by the same research group [21,23].

### Table 1

Experimental data for FO mode experiments (with no applied pressure) with DI water feed and various NaCl draw solution concentrations at 20 °C. K values were determined by fitting water flux data to Eq. (6). S values were calculated using Eq. (3) with NaCl diffusivity of $1.33 \times 10^{-5}$ m$^2$/s.

<table>
<thead>
<tr>
<th>Draw (NaCl) solution concentration (M)</th>
<th>Feed solution concentration (M)</th>
<th>Water flux ($\mu$m/s)</th>
<th>Solute resistance to diffusion, K ($10^9$ s/m)</th>
<th>Structural parameter, S ($10^{-3}$ m)</th>
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<td>Average</td>
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<td>3.85 ± 0.31</td>
<td>3.52 ± 0.15</td>
<td>512 ± 40.96</td>
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</tbody>
</table>

### Table 2

Experimental PAO data for FO mode experiments (with applied pressure as indicated) with DI water feed and various NaCl draw solution concentrations at 20 °C. K values were determined by fitting water flux data to Eq. (6). S values were calculated using Eq. (3) with NaCl diffusivity of $1.33 \times 10^{-5}$ m$^2$/s.

<table>
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<th>Draw (NaCl) solution concentration (M)</th>
<th>Applied hydraulic pressure (bar)</th>
<th>Water flux ($\mu$m/s)</th>
<th>Solute resistance to diffusion, K ($10^9$ s/m)</th>
<th>Average K</th>
<th>Structural parameter, S ($10^{-3}$ m)</th>
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### 4.2. Flux behavior of PAO in PRO mode

The results of PAO experiments in PRO mode are shown in Fig. 4 and summarized in Table 3 along with the corresponding mass transfer coefficients in the draw solution channel as obtained from fitting the water flux data to Eq. (7). Note that all experimental conditions were identical to the previously described PAO runs in FO mode (Section 4.1), except the membrane orientation.

The water fluxes in PRO mode (Fig. 4, Table 3) were higher compared to FO mode (Fig. 3, Table 2) at all draw solution concentrations under no hydraulic pressure. This observation is attributed to the insignificant ICP in PRO mode when the feed DI water is at the porous support layer side of the membrane [1,2]. Under applied hydraulic pressure, the measured water flux was lower than model prediction. Regardless of draw solution concentration, the water fluxes at an applied pressure of 3.45 bar were always lower than the water fluxes at no applied pressure. At 2.0 M draw solution concentration, the water flux at 13.8 bar was even lower than the water flux at no applied pressure. Although the driving force in PAO increased with increasing the applied hydraulic pressure in both FO and PRO modes, the water flux behaviors were quite different. We discuss the mechanisms governing the flux behavior in the next section.

### 4.3. Mechanisms of flux behavior in PAO

To explain the abnormal water flux behavior in PRO mode, we first considered the ‘shadow effect’ [16]. The shadow describes the
Together with the reduction in the effective membrane surface as described above, the impact of changes in the cross flow rate on the draw solution side was also considered. As the applied pressure increased in PRO mode, the flow rate decreased from 1 l/min to approximately 0.6 l/min. This decrease in flow rate is attributed to the reduction in the flow channel effective height of the membrane test cell caused by the applied hydraulic pressure. The mass transfer coefficient is related to the Reynolds number \((Re = d_0 U_0/\nu)\), where \(d_0\) is the hydraulic diameter, \(U_0\) is the average cross flow velocity, and \(\nu\) is the kinematic viscosity. If the cross flow velocity is reduced, the mass transfer coefficient \((k)\) decreases, which enhances ECP, and thus may affect water flux as shown in Eq. (7).

To examine the effect of flow rate reduction on the draw solution side, a set of experiments were conducted, while increasing the output of the gear pump to maintain the flow rate constant regardless of the applied hydraulic pressure. Although the results are not shown here, identical or slightly increased waters flux were observed, suggesting that the effect of flow rate change in the channel is almost negligible. Accordingly, the reduction in the effective membrane surface area that is in contact with the draw solution by the spacers under the action of the applied hydraulic pressure – the “chemical spacer effect” – is likely the dominant mechanism for the water flux behavior in PAO.

### 4.4. Reverse solute flux and reverse flux selectivity behavior in PAO

Reducing reverse solute flux is one of the main challenges in FO systems. The solutes passing through the membrane from the draw side to feed side reduce effective the osmotic pressure between draw and feed sides and necessitate the need to supplement the draw solution, all of which adversely impact FO performance. Moreover, the reverse solute flux can contribute to fouling on the membrane through cake-enhanced osmotic pressure [19,24]. The driving force for reverse solute flux in FO is the draw solute concentration difference across the active layer of the membrane, which is also the driving force for water permeation. Accordingly, the reverse solute flux in PAO is expected to decrease with increasing applied hydraulic pressure. This is because the permeate water by the applied hydraulic pressure decreases the effective osmotic pressure and thus reduces the solute concentration difference across the membrane. Fig. 7 illustrates the changes in the concentration profiles and the impact of pressure (i.e., operation in PAO) on ICP and ECP in both FO and PRO modes.

Fig. 8(a) and (b) shows the reverse solute flux for both the FO and PRO membrane orientations. As expected, the reverse solute flux dramatically decreases after applying hydraulic pressure. In addition, the reverse solute flux in FO mode is lower than in PRO mode. Fig. 8(c) and (d) shows the reverse flux selectivity \((J_w/J_s)\), which characterizes the membrane selectivity toward the draw solution [21,22]. As shown in Fig. 8(c) and (d), the reverse flux selectivity values were similar when no pressure was applied, but increased with increasing pressure. Note that the reverse flux selectivity varies with the applied hydraulic pressure and the concentration of draw solute, as well as the membrane orientation. Accordingly, the optimal operating conditions for PAO may be determined by simultaneously considering water flux and reverse solute flux.

### 4.5. Potential application of PAO

In order to verify the effectiveness of PAO, we introduce the water flux improvement ratio which is defined as

\[
\text{Improvement Ratio (IR)} = \frac{J_{w,PAO}}{J_{w,FO}}
\]

where \(J_{w,FO}\) is the water flux at non-pressure condition (i.e. FO) and \(J_{w,PAO}\) is water flux at a certain applied pressure on feed the side.
Fig. 9 compares the water flux improvement ratios at the various applied hydraulic pressures and draw solute concentrations for the FO and PRO membrane orientations. At low draw solution concentration (i.e. 0.5 M), PAO is effective in enhancing the permeate water flux, particularly in FO mode. As the draw solution concentration increases, the increasing rate of the water flux improvement ratio decreases. Notably, the impact of pressure on increasing the water flux was much more dramatic in FO mode.
compared to PRO mode. The increase in PAO water flux in FO mode under the condition of low draw solution concentration and the concomitant decrease in reverse solute flux suggest potential PAO potential applications as the one discussed below.

Recently, synergistically coupling FO with RO has attracted increasing interest for augmenting water supply. In this system, the FO membrane is applied between seawater (as draw solution) and wastewater effluent (as feed solution), which reduces the salinity and osmotic pressure of the seawater feed to RO desalination [25–28]. Fig. 10(a) depicts an example of such systems: a 2-stage FO with RO system for seawater desalination. One of the limitations posed by such FO–RO systems is low FO water flux and thus low recovery in the FO process. The recovery of the FO–RO system may be increased by adopting PAO, which can increase water flux by additional hydraulic pressure. Fig. 10(b) illustrates FO–PAO–RO system, in which a second stage FO is replaced with PAO, allowing for higher water flux and recovery than the simple 2-stages FO with RO. As shown in Fig. 9, PAO is more effective when the concentration of draw solution is low, such as the case in the second stage of FO. Using PAO at that stage, the seawater is further diluted, leading to lower energy consumption for RO desalination process. Furthermore, control of the driving force, the hydraulic pressure, is easier in PAO than FO, leading to flexibility in system design and operation. The required pressure for the PAO process may be obtained from the high pressure of the RO brine via the pressure exchanger (PX), implying that there is no need to use external energy source. Further studies on the technical and economic feasibility of PAO process are needed to validate this concept.

Fig. 8. Reverse solute flux and reverse flux selectivity for the PAO experiments depicted in Figs. 3 and 4: (a) Reverse solute flux in FO mode, (b) reverse solute flux in PRO mode, (c) reverse flux selectivity in FO mode, and (d) reverse flux selectivity in PRO mode.

Fig. 9. Water flux improvement ratio in PAO for the various applied hydraulic pressures and draw solution concentrations: (a) FO mode and (b) PRO mode. Data are from the experiments described in Figs. 3 and 4.
5. Conclusion

In this work, PAO system was systematically investigated as an FO technology to overcome the limitations of conventional FO: low water flux and high reverse solute flux. The measured water flux in PAO showed relatively good agreement with model prediction in FO mode. The water flux increased linearly with increasing applied hydraulic pressure, allowing high water flux without increasing draw solution concentration. On the other hand, the measured water flux in PRO mode was much lower than model predictions. This observation is attributed to the reduction in the effective draw solution concentration. On the other hand, the measured water flux in PRO mode was much lower than model predictions. This is because the convection of water originating from the addition of the applied hydraulic pressure reduced concentration difference across the membrane, which is a main driving force for reverse solute flux. When considering water flux and reverse solute flux, PAO is effective in FO mode under the condition of low draw solution concentration. PAO seems to have potential to be used in an FO–RO hybrid system, especially when it is designed for high recovery of water.

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