Desalination by ammonia–carbon dioxide forward osmosis: Influence of draw and feed solution concentrations on process performance

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Abstract

Forward (direct) osmosis (FO) using semi-permeable polymeric membranes may be a viable alternative to reverse osmosis as a lower cost and more environmentally friendly desalination technology. The driving force in the described FO process is provided by a draw solution comprising highly soluble gases—ammonia and carbon dioxide. Using a commercially available FO membrane, experiments conducted in a crossflow, flat-sheet membrane filtration cell yielded water fluxes ranging from 1 to 10 \text{ gpd ft}^{-2} \text{ d}^{-1} \text{ or 3.6 to 36.0 l m}^{-2} \text{ h}^{-1} for a wide range of draw and feed solution concentrations. It was found, however, that the experimental water fluxes were far lower than those anticipated based on available bulk osmotic pressure difference and membrane pure water permeability data. Internal concentration polarization was determined to be the major cause for the lower than expected water flux by analysis of the available water flux data and SEM images of the membrane displaying a porous support layer. Draw solution concentration was found to play a key role in this phenomenon. Sodium chloride rejection was determined to be 95–99% for most tests, with higher rejections occurring under higher water flux conditions. Desalination of very high sodium chloride feed solutions (simulating 75% recovery of seawater) was also deemed possible, leading to the possibility of brine discharge minimization.

1. Introduction

A worldwide shortage of unimpaired freshwater has forced the tapping of saline water sources in water-starved countries. Current desalination technologies, such as reverse osmosis (RO), are, however, expensive and energy intensive. Limited recovery, typically 35–50% for seawater [1], is another drawback of RO. The remaining liquid, now concentrated brine, is discharged into the ocean. This is a critical environmental drawback to RO and limits its use to coastal areas since brine from brackish ground-water desalination cannot be disposed of inland in an economical manner.

Forward (or direct) osmosis (FO) is a technology that may be able to desalinate saline water sources at a reduced cost and at high recovery. In forward osmosis, like RO, water naturally transports across a semi-permeable membrane that is impermeable to salt. However, instead of using hydraulic pressure to create the driving force for water transport through the membrane, the FO process utilizes an osmotic pressure gradient. A “draw” solution having a significantly higher osmotic pressure than the saline feed water flows against the permeate side of the membrane, and water naturally transports across the membrane by osmosis. The osmotic driving forces in FO can be significantly greater than hydraulic driving forces in RO, potentially leading to higher water flux rates and recoveries. The lack of hydraulic pressure may make the process less expensive than RO, while the minimization of brine discharge reduces the environmental impact of the desalination process.

Previous work on osmosis through semi-permeable membranes, while limited, still exposed the two significant drawbacks of FO. Membrane technology has advanced around the prospect of RO, not FO. Research conducted on osmotic processes concluded that RO membranes were not ideal for osmosis as water flux was found to be low [2–5]. Finding easily separable draw solutes is the second drawback of FO. Several patents describe different configurations for the use and removal of FO draw solutes [6–14]. Choosing a tailored FO membrane as well as an appropriate draw solute is critical in making the desalination...
process feasible. In the work presented in this paper, a membrane specifically designed for FO was used and our draw solutes and solute composition were chosen based on several important chemical attributes.

The ideal draw solute has several characteristics. The solute(s) must have a high osmotic efficiency, namely high solubility in water and relatively low molecular weight, which can lead to high osmotic pressures. The solute must also be easily and inexpensively separated to yield potable water, without being consumed in the process. We have recently demonstrated that these criteria are satisfied by using a draw solution of highly soluble gases—ammonia (NH₃) and carbon dioxide (CO₂) [15]. These gases are highly soluble in water and have a low molecular weight while also being relatively easy to remove from water. We have shown that by proper management of ammonia and carbon dioxide ratios and ammonium salt speciation, we can generate very high osmotic driving forces [16]. Upon moderate heating (near 60°C) [17], the ammonium draw solutes decompose into ammonia and carbon dioxide gases that can be separated by standard means. The separated gases can then be used to regenerate the draw solution. This draw solution separation step in the FO desalination process will not be addressed in this paper.

The objective of this paper is to investigate the relationships among the draw solution concentration, feed water salinity, and permeate flux behavior in the ammonia–carbon dioxide FO process. The ammonia–carbon dioxide draw solution concentration is varied over a wide range (up to 6 M), processing feed waters with salinities as high as 2 M sodium chloride (NaCl). On the basis of these results, the mechanisms governing the permeate water flux behavior in the ammonia–carbon dioxide FO process are elucidated and discussed.

2. Materials and methods

2.1. Ammonia–carbon dioxide draw solution and sodium chloride feed solution

The draw solution is made by mixing ammonium bicarbonate (NH₄HCO₃) and ammonium hydroxide (NH₃OH) with deionized water at proper proportions to produce a solution of the desired concentration of ammonium salts [15,16]. Higher concentrations of ammonia and carbon dioxide salts require a higher ratio of ammonia to carbon dioxide, which favors the formation of ammonium carbamate, a highly soluble ammonium salt. The ammonia to carbon dioxide molar ratios used ranged from 1.2 for the 1.1 M draw solution to 1.4 for the 6 M draw solution.

The feed solution was composed of NaCl dissolved in deionized water. Concentrations ranged from 0.05 to 2 M. The 2 M NaCl concentration is indicative of 75% recovery of a 0.5 M NaCl solution, which is close to an initial seawater concentration. The temperature of the feed solution was 50°C.

2.2. Forward osmosis membrane

The membrane tested in this work was provided by Hydration Technologies Inc. (Albany, OR). The proprietary FO membrane, denoted as CA in this paper, is thought to be made from a hydrophilic cellulose-based polymer [13]. Fig. 1 shows a cross-sectional view of the membrane taken by SEM, indicating that the thickness of the membrane is less than 50 μm. It is evident that the structure of the CA membrane is quite different from standard RO membranes. RO membranes typically consist of a very thin active layer (less than 1 μm) supported by a much thicker porous polymer supporting layer [18]. These layers are mechanically supported by a fabric support layer. Fig. 1 indicates that the CA FO membrane lacks any thick fabric layer. An embedded polyester mesh provides the mechanical support normally provided by the thick fabric layer [13].

2.3. Forward osmosis crossflow set-up

Fig. 2 depicts the apparatus employed in our laboratory-scale FO experiments. The crossflow membrane unit is a SEPA cell (GE Osmonics, Trevose, PA), modified to have channels on both sides of the membrane. The ammonia–carbon dioxide draw solution is flowing on the permeate side and the NaCl solution on the feed (active layer) side. Co-current flow is used to reduce strain on the suspended membrane. Mesh spacers are inserted within both channels to improve support. The spacers also promote turbulence and mass transport. Variable speed peristaltic pumps (Manostat, Barrington, IL) are used to pump the liquids. A constant temperature water bath (Neslab, Newington, NH) is used to maintain the feed solution temperature, whereas the draw solution temperature is kept constant by a heating mantle (Glas-Col, Terre Haute, IN). Both the feed and draw solutions were held at the same temperature (50°C) during the FO tests and were controlled to within ±1°C.

2.4. Water flux and salt rejection measurements

Water flux is determined by measuring the weight change (Ohaus, Pine Brook, NJ) of the draw solution over a selected time period at the initial stage of the process. As water transports by osmosis across the membrane from the saline feed water
into the draw solution, the draw solution weight increases. The rate at which the volume increases (determined from the weight change) divided by the membrane area yields the water flux.

For determining NaCl rejection, a sample of draw solution is taken after a complete FO run and is boiled until all water evaporates. During the boiling process, the dissolved ammonium salts decompose into NH₃ and CO₂ gases. The remaining solid NaCl is hydrated with a known volume of deionized water, and the NaCl concentration is measured by a chloride selective electrode (Cole-Parmer, Vernon Hills, IL). Based on the amount of water passed into the draw solution over the course of the experiment and the final amount of NaCl in the draw solution, the permeate NaCl concentration is determined. The percent salt rejection, $R$, is then calculated from

$$R = \frac{100}{1 - \frac{C_p}{C_F}}$$

where $C_p$ and $C_F$ are the permeate and feed NaCl concentrations, respectively.

2.5. Dependence of membrane water permeability on feed NaCl concentration

Osmotic permeability measurements to determine whether the water permeability coefficient changes at high feed NaCl concentrations were taken with a redesigned FO cell having well-controlled hydrodynamics. The crossflow cell is of plate-and-frame design with a rectangular channel on each side of the membrane. The channel has dimensions of 7.7 cm length, 2.6 cm width, and 0.3 cm height, providing an effective membrane area of 20.0 cm². The crossflow velocity during the testing was 21.4 cm/s. An RO cell with similar dimensions was used to determine the pure water hydraulic permeability for comparison.

To conduct the osmotic permeability measurements, NaCl solution was used on the active layer side of the membrane, while deionized water ran along the backing (support) layer side. Osmotic flux data were collected for NaCl concentrations ranging from 0.05 to 2 M, corresponding to the feed solutions that were used in the FO experiments described above. The temperature of both solutions was maintained at 50°C, which was the temperature used for the FO and pure water permeability experiments.

High driving forces and correspondingly high water fluxes can cause severe concentration polarization effects, which occur on the permeate side of the membrane (in this case, the active layer). To account for this phenomenon, the extent of concentration polarization was calculated from film theory. The Sherwood number, $Sh$, was first determined by using either the laminar or turbulent flow correlation for a rectangular channel [19,20]:

$$Sh = 1.85 \left( Re Sc \frac{d_h}{L} \right)^{0.33}$$

$$Sh = 0.04 Re^{0.8} Sc^{0.33}$$

where $Re$ is the Reynolds number, $Sc$ the Schmidt number, $d_h$ the hydraulic diameter, and $L$ is the length of the channel. By...
concentrations

3.1. Water flux with variable feed and draw solution concentrations

Water flux across a semi-permeable membrane when osmotic pressure is the driving force is given by [19,23]:

\[ J = A \sigma \Delta \pi \]  

(6)

Here, \( A \) is the water permeability coefficient, \( \sigma \) the reflection coefficient, and \( \Delta \pi \) is the difference in osmotic pressures across the membrane between the draw and feed solution at the separating interface (i.e., at the membrane active layer). The reflection coefficient approaches 1.0 for a completely rejecting membrane. Because the rejection in most of our FO experiments was above 95\% (to be discussed later in the paper), the reflection coefficient for the FO runs presented in this paper was assumed to be unity. It should be noted, however, that determination of the reflection coefficient for our system involving ammonia–carbon dioxide as a draw solution and NaCl as a feed solution may be complicated by the leakage of free ammonia to the feed size of the membrane. Because our experiments involved numerous combinations of draw and feed solution concentrations, the determination of the reflection coefficient for each individual run was not feasible.

The bulk osmotic pressures of our draw (NH\(_3/CO_2\)) and feed (NaCl) solutions were calculated using software from OLI Systems Inc. (Morris Plains, NJ) and Aspen HYSYS® (Cambridge, MA). The OLI Systems software uses thermodynamic modeling based on published experimental data to predict the properties of solutions over a wide range of concentrations and temperatures. The molar concentrations of the added ammonium solutes – ammonium bicarbonate (NH\(_4\)HCO\(_3\)) and ammonium hydroxide (NH\(_4\)OH) – for each draw solution were used to calculate the osmotic pressure. The OLI software was also used to calculate the speciation and pH of the draw solutions. The pH of the draw solutions ranged from 7.73 for the 1.1 M draw solution to 8.26 for the 6 M draw solution.

Our previous work on the ammonia–carbon dioxide FO desalination process [15] discussed limited water flux results for a single feed solution concentration. In this investigation we examine the permeate water flux behavior over a wide range of feed as well as draw solution concentrations. Fig. 3 presents permeate water flux data for different feed NaCl concentrations as a function of the osmotic pressure difference, \( \pi_D - \pi_F \), where \( \pi_D \) and \( \pi_F \) are the bulk osmotic pressures of the draw and feed solutions, respectively.

Assuming the water permeability coefficient, \( A \), is constant in all tests and \( \sigma \approx 1 \), Eq. (6) would predict a proportional increase in flux with increasing osmotic driving force, regardless of feed solution concentration. Fig. 3, however, suggests that feed solution concentration may be a key parameter that influences water flux in the FO process. The data in Fig. 3 indicate that for similar differences in osmotic pressure between the bulk feed and draw solutions (\( \pi_D - \pi_F \)), higher concentration feed solutions produce less flux. We must consider, however, that higher concentration feed solutions require more concentrated draw solutions to achieve the same theoretical osmotic driving force. This phenomenon, therefore, may not be a feed salinity effect. The draw solution may also play a role.

Assuming that the full osmotic driving force, referred to as a theoretical driving force, can be realized, a theoretical water flux can be calculated based on Eq. (6). We use a constant water permeability coefficient value of \( 5.69 \times 10^{-12} \text{ mPa}^{-1} \text{s}^{-1} \) in these calculations, determined by reverse osmosis tests of the CA membrane at 50°C. Later in this paper, we will reassess this
Table 1

Water flux, performance ratio, and NaCl rejection results for the FO runs carried out in this study under the stated feed and draw solution concentrations (and the corresponding osmotic pressures)

<table>
<thead>
<tr>
<th>Feed concentration (M)</th>
<th>Draw concentration (M)</th>
<th>( \pi_F ) (atm)</th>
<th>( \pi_D ) (atm)</th>
<th>( \Delta \pi ) (atm)</th>
<th>Theoretical flux in ( \mu \text{m}^3/\text{cm}^2 \text{h} )</th>
<th>Experimental flux in ( \mu \text{m}^3/\text{cm}^2 \text{h} )</th>
<th>Performance ratio</th>
<th>Rejection (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.05</td>
<td>2.5</td>
<td>47.4</td>
<td>44.9</td>
<td>25.8</td>
<td>5.3 (11.2)</td>
<td>0.204</td>
<td>97.90</td>
<td></td>
</tr>
<tr>
<td>0.05</td>
<td>3</td>
<td>127.6</td>
<td>125.1</td>
<td>72.0</td>
<td>10.1 (21.4)</td>
<td>0.120</td>
<td>99.45</td>
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</tr>
<tr>
<td>0.2</td>
<td>9.8</td>
<td>47.4</td>
<td>37.6</td>
<td>21.6</td>
<td>4.1 (8.7)</td>
<td>0.188</td>
<td>93.72</td>
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<tr>
<td>0.2</td>
<td>9.8</td>
<td>127.6</td>
<td>117.8</td>
<td>67.8</td>
<td>6.8 (14.5)</td>
<td>0.101</td>
<td>97.34</td>
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<td>0.2</td>
<td>9.8</td>
<td>249.5</td>
<td>228.8</td>
<td>131.7</td>
<td>9.4 (19.9)</td>
<td>0.068</td>
<td>98.12</td>
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<tr>
<td>0.5</td>
<td>24.8</td>
<td>47.4</td>
<td>22.6</td>
<td>13.0</td>
<td>0.9 (2.0)</td>
<td>0.074</td>
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<td>24.8</td>
<td>73.3</td>
<td>48.4</td>
<td>27.9</td>
<td>2.8 (6.0)</td>
<td>0.102</td>
<td>96.05</td>
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<tr>
<td>0.5</td>
<td>24.8</td>
<td>94.5</td>
<td>69.7</td>
<td>40.1</td>
<td>3.1 (6.6)</td>
<td>0.078</td>
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<tr>
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<td>127.6</td>
<td>102.8</td>
<td>59.2</td>
<td>4.6 (9.7)</td>
<td>0.077</td>
<td>98.07</td>
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<tr>
<td>0.5</td>
<td>24.8</td>
<td>193.3</td>
<td>168.5</td>
<td>97.0</td>
<td>5.9 (12.5)</td>
<td>0.061</td>
<td>97.14</td>
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<tr>
<td>0.5</td>
<td>24.8</td>
<td>249.5</td>
<td>213.8</td>
<td>123.1</td>
<td>6.4 (13.5)</td>
<td>0.049</td>
<td>98.70</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>51.5</td>
<td>127.6</td>
<td>76.1</td>
<td>43.8</td>
<td>2.3 (4.8)</td>
<td>0.052</td>
<td>93.89</td>
<td></td>
</tr>
<tr>
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<td>193.3</td>
<td>141.8</td>
<td>81.6</td>
<td>4.4 (9.4)</td>
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<tr>
<td>2</td>
<td>113.8</td>
<td>193.3</td>
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<td>45.8</td>
<td>1.1 (2.3)</td>
<td>0.024</td>
<td>90.14</td>
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<tr>
<td>2</td>
<td>113.8</td>
<td>249.5</td>
<td>124.9</td>
<td>73.5</td>
<td>2.0 (4.3)</td>
<td>0.026</td>
<td>96.85</td>
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</table>

It is also assumed that \( \sigma \approx 1 \) as discussed earlier in the paper. In all experiments presented in Fig. 3, the measured water flux was lower than the theoretical flux. We define the performance ratio as the experimental water flux divided by the theoretical water flux. This ratio is equivalent to the percentage of the bulk osmotic pressure difference that is effectively generating water flux across the FO membrane.

Table 1 summarizes the FO experimental data, including the feed and draw solution concentrations used, the corresponding osmotic pressures and the osmotic pressure difference (\( \Delta \pi \)), the calculated (theoretical) water flux based on the osmotic pressure difference, the measured water flux, and the performance ratio. Also included in this table are salt rejection data, to be discussed later. The variation of the performance ratio with draw solution concentration for the different feed solution concentrations is shown in Fig. 4. The results demonstrate that the performance ratio decreases with increasing draw and feed solution concentrations. At best, around 20% of the osmotic pressure driving force is utilized, while at higher draw solution concentrations, as little as 5% or less of the osmotic pressure driving force is realized for the experimental conditions discussed here.

3.2. Processes governing the permeate flux behavior

The much lower than expected water flux in the previously described experiments is likely caused by several membrane associated transport phenomena. Specifically, two types of concentration polarization (CP) phenomena – external CP and internal CP – can take place in FO as discussed below. In addition, exposure of the membrane to very concentrated salt solutions such as those used in our FO runs may influence the water permeability of the membrane. These possible causes are analyzed below.

3.2.1. External concentration polarization

In pressure-driven membrane processes, convective permeate flow causes a buildup of solute at the membrane active layer surface. Referred to as CP, this reduces permeate water flux due to increased osmotic pressure that must be overcome with hydraulic pressure. Permeate convection, and hence CP, is not limited to pressure-driven membrane processes and also occurs in FO on the membrane feed side. This may in part explain the low performance ratios in Fig. 4. A similar but dilutive effect occurs on the permeate (support layer) side, as permeating water dilutes the draw solution along the external membrane backing layer surface. Higher concentration draw solutions will increase the severity of this “dilutive” CP, since higher draw solution concentrations yield higher water flux rates. Combined, the “concentrative” and dilutive external CP phenomena reduce the effect of draw solution concentration on the performance ratio for various feed solution concentrations. Experimental conditions: crossflow rate and temperature of both feed and draw solution of 30 cm/s and 50 °C, respectively.

Fig. 4. The effect of draw solution concentration on the performance ratio for various feed solution concentrations.
the effective osmotic driving force. In a scaled up process, the effects of the external CP phenomenon on driving force can be minimized by using crossflow filtration with hydrodynamics designed to produce sufficient shear and turbulence at the membrane surface [27].

Our system (SEPA cell) incorporates a shallow channel (0.864 mm) and mesh spacers, minimizing both concentrative and dilutive external CP effects through high shear and increased turbulence. For the experimental conditions of our FO runs (crossflow velocity of 30 cm/s and a temperature of 50 °C), the concentration polarization modulus values were calculated from film theory for the feed side of the membrane. Reynolds numbers in the system ranged from about 940 to 960 (the range is due to viscosity and density variability over the range of NaCl concentrations).

We calculated the mass transfer coefficients and the CP moduli for each of our tests based on Eqs (2), (4), and (5). Note that for this case, Eq (5) has a positive exponent. We did not use the turbulent correlation because the flow in this case is in the laminar regime. CP moduli for the various feed solutions and water fluxes ranged from 1.03 to 1.33, meaning that effective membrane surface concentration was not much greater than the bulk concentration. Since the feed NaCl concentration was always significantly less than the draw solution concentration, small increases in the NaCl surface concentration at the active layer did not critically affect overall driving force. It is also important to note that the higher CP moduli were associated with the lower feed NaCl concentrations (due to higher water fluxes), further mitigating the effect of external CP on the driving force. External CP on the feed side of the membrane, therefore, can be ruled out as the cause for the markedly lower than expected water flux.

Dilutive external CP (i.e., on the outer support layer side) was not considered since the water permeating the backing layer of the membrane contained some concentration of the draw solute. This permeate concentration would mitigate the polarization effects on the backing side of the membrane since pure water is not entering the bulk solution at the membrane surface. Polarization effects on the permeate side of the active layer within the porous support layer are considered in the next section.

3.2.2. Internal concentration polarization

When an osmotic pressure gradient is established across a completely rejecting dense symmetric membrane, as depicted in Fig. 5a, the driving force is the difference in osmotic pressures of the bulk solutions in the absence of concentrative and dilutive external CP. When a composite or asymmetric membrane consisting of a dense separating layer and a porous support layer is used in forward osmosis, two phenomena can occur depending on the membrane orientation.

If the porous backing layer of an asymmetric membrane is facing the feed solution, a polarized layer is established along the inside of the dense active layer as water and solute propagate the porous support layer. Referred to as internal concentration polarization [3,4], this phenomenon, illustrated in Fig. 5b, is

![Diagram](image-url)
similar to concentrative external CP, except that it takes place within the protected confines of the porous backing layer, and hence cannot be minimized by crossflow.

Our system, however, is run with the membrane in the opposite orientation. In our FO configuration, the NaCl feed is channeled on the active layer side of the membrane and the ammonia–carbon dioxide draw solution is channeled on the backing layer side. This was done to most closely mimic the typical membrane desalination configuration where the feed is directed against the active layer. Upon contact with the backing layer of the membrane, the draw solution will diffuse into the porous substructure. The osmotic driving force between the feed and draw solutions will then be established across the dense active layer of the membrane. As water permeates the active layer, the draw solution within the porous substructure becomes diluted. We refer to this as dilutive internal CP. As with concentrative internal CP, dilutive internal CP cannot be controlled by crossflow. The unmitigated dilutive internal CP phenomenon significantly decreases the net driving force, leading to decreased flux. Illustrated in Fig. 5c, this phenomenon occurs when asymmetric or thin film composite membranes are used in an FO mode with the draw solution against the porous support/backing layer.

For dilutive internal CP to occur in our tests, the CA membrane would have to contain a porous support layer. Fig. 1 does not reveal any obvious asymmetry within the CA membrane. If there were no porous support layer, then no dilutive internal CP would occur. However, a closer inspection of the membrane, shown in Fig. 6, uncovers asymmetry. Fig. 6a shows a dense active layer, while Fig. 6b reveals a porous supporting structure. This porous structure should result in substantial dilutive internal CP that explains the much lower than expected water flux. Additional analysis of the marked effect of dilutive internal CP in our system is given later in this paper.

3.2.3. Does the membrane water permeability coefficient change with feed NaCl concentration?

Knowing the membrane water permeability at a given set of system conditions is critical to calculating the theoretical flux and performance ratio as well as eliminating uncertainty in the cause of various flux behaviors. Previous studies on osmosis through asymmetric cellulose acetate membranes have reported that membrane water permeability is often found to be significantly lower than the corresponding hydraulic water permeability determined by RO [28–32]. Pusch [32] presented data showing a precipitous drop in water permeability as the membrane became exposed to more salt. Much of the data, however, was in the form of observations of apparent permeability based on water flux and bulk solution driving force rather than the true membrane permeability. This behavior was attributed to internal concentration polarization effects within the porous support layer. A few studies, however, suggested that high salt concentrations would actually lower the intrinsic water permeability of a membrane through osmotic deswelling or other related phenomena [29,30]. Our performance ratio data was calculated using the hydraulic water permeability coefficient obtained for pure water with RO at 50°C. In order to test the influence of NaCl feed solution on the water permeability coefficient, we carried out osmotic permeability runs exposing the active layer to increasing NaCl feed concentrations (up to 2 M) while DI water was placed against the support layer.

Fig. 7 shows the flux data gathered, based on the experimental procedure outlined in Section 2.5. As the NaCl bulk solution concentration is increased, the flux behavior is not linear. This may suggest that the permeability of the membrane may be changing at higher NaCl concentrations. The water fluxes, however, are quite high for the tests at high NaCl concentrations where nonlinearity is obvious, suggesting that dilutive external CP on the permeate side of the membrane may significantly reduce the osmotic driving force.

To test our assumption that dilutive external CP rather than reduction in membrane water permeability is the cause for the deviation from linearity, we carried out CP calculations as outlined in Section 2.5. The Reynolds number was found to be around 2100 for the flow conditions in the new FO cell described above. With a Reynolds number on the cusp of the laminar flow regime, coupled with the mesh spacers, transition flow is likely occurring. We therefore used Eqs. (2)–(5) to account for this phenomenon and calculated the NaCl concentration at the membrane surface using the average of the turbulent and laminar CP modulus for each test [22]. The effective osmotic driving force is
Fig. 7. Flux data with NaCl draw solution on the active layer and deionized water on the backing layer. Experiments were carried out with a specially designed FO cell described in Section 2.5. Experimental conditions: crossflow velocity and temperature of both feed and draw solution of 21.4 cm/s and 50 °C, respectively. Note that a water flux of 10 μm/s corresponds to 21.2 gal ft$^{-2}$ d$^{-1}$ (gfd) or 36.0 l m$^{-2}$ h$^{-1}$.

then calculated using the OLI software as before. This corrected data is presented in Fig. 8.

Fig. 8 shows that flux increases linearly with increasing osmotic pressure at the membrane surface when the concentration at the membrane surface is calculated using film theory. Also plotted on Fig. 8 is the hydraulic permeability data taken for the CA membrane using RO. The osmotic flux data matches closely to the hydraulic flux data, allowing us to conclude that the water permeability coefficient of our CA membrane does not change when exposed to the NaCl feed concentrations used in the FO runs (summarized in Table 1). This confirms that changes in membrane permeability are not affecting water flux.

Fig. 9. Flux data (for the FO experiments summarized in Table 1) plotted against the logarithm of the ratio of feed and draw solution osmotic pressures. Experimental conditions: SEPA cell with crossflow rate and temperature of both feed and draw solution of 30 cm/s and 50 °C, respectively. Note that a water flux of 10 μm/s corresponds to 21.2 gal ft$^{-2}$ d$^{-1}$ (gfd) or 36.0 l m$^{-2}$ h$^{-1}$.

3.3. Water flux uniquely depends on a normalized osmotic pressure driving force

Another way of evaluating the flux behavior and confirming the presence of dilutive internal CP is by normalizing the driving force. Two methods are presented here. The first, suggested by Loeb et al. [5], considers the ratio of draw solution to feed solution bulk osmotic pressure, π$_D$/π$_F$. It was shown that the effective driving force in a forward osmotic process that is influenced by internal CP can be approximated by ln(π$_D$/π$_F$). The data from Table 1 are normalized using this method and shown in Fig. 9. The linear trend confirms that our data correspond well with such analysis of flux behavior in an osmotic process through an asymmetric RO membrane.

To better conceptualize how the concentration of draw solution affects dilutive internal CP, we use another normalization technique. By plotting our water flux against the bulk osmotic pressure difference normalized by the feed osmotic pressure ($\pi_D - \pi_F)/\pi_F$, a strong logarithmic trend is apparent (Fig. 10). This observation supports our conclusion that the water flux behavior in governed by dilutive internal CP. At low normalized driving forces, flux increases sharply with increasing driving force. However, at higher driving forces caused by higher draw solution concentrations, flux increases diminish as driving force increases. High draw solution concentrations generate larger osmotic driving forces which produce more water flux. Higher water fluxes increase the severity of dilutive internal CP by increasing the degree of dilution within the backing layer of the membrane, lowering the performance ratio and diminishing the increase in flux for higher draw solution concentrations.

3.4. Salt rejection and recovery

Even with dilutive internal CP, water flux is still reasonably high. For an efficient FO desalination process, it is also imperative that salt rejection be high. Rejection data for all our...
Fig. 10. Flux data (for the FO experiments summarized in Table 1) plotted against the normalized osmotic pressure difference. Note that $\pi_D$ is the bulk osmotic pressure of the draw solution and $\pi_F$ is the osmotic pressure of the feed solution.

Experimental conditions: SEPA cell with crossflow rate and temperature of both feed and draw solution of 30 cm/s and 50 $^\circ$C, respectively. Note that a water flux of 10 $\mu$m/s corresponds to 21.2 gal ft$^{-2}$ d$^{-1}$ (gfd) or 36.0 l m$^{-2}$ h$^{-1}$.

experiments summarized in Table 1 are presented in Fig. 11. The results indicate that NaCl rejection is above 95% for most of the experiments, and approaches 99% for some of the higher water flux experiments. Rejection increases with increasing water flux due to the “dilution effect”, which occurs when water flux is increased without a subsequent increase in salt flux. Increasing the osmotic pressure driving force only affects water flux and not salt flux, and rejection is thereby improved.

4. Concluding remarks

The data presented in this paper demonstrate that the draw solution concentration has a significant effect on the flux behavior of the ammonia–carbon dioxide FO desalination process. It was determined that dilutive internal concentration polarization is primarily responsible for a much lower than expected water flux. Data showed that increasing the draw solution concentration leads to a decrease in the percentage utilization of the available bulk osmotic driving force, or a decrease in performance ratio, due to an increase in the severity of the dilutive internal CP. This was concluded after external CP and possible osmotic deswelling of the FO membrane were shown to have negligible effects on FO performance. The performance data show that in spite of dilutive internal CP, water flux is still reasonably high with the FO membrane. The FO system was shown to perform well in terms of NaCl rejection and to operate at very high feed concentrations, simulating operation at high feedwater recovery. While overall performance of the FO process was satisfactory, the FO membrane tested in this study is not optimal for this process since its primary use is not for desalination. The development of a semi-permeable membrane having high salt rejection and minimal internal CP will lead to improved performance of this forward osmosis desalination process.

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