1. Introduction

Forward osmosis (FO), also known as direct osmosis, is a rapidly growing field in membrane science. Although it has not been studied as extensively as pressure-driven membrane processes (e.g. reverse osmosis (RO), nanofiltration, and ultrafiltration), FO was first considered for desalination and osmotic power production over 40 years ago [1–3]. Throughout the last decade, FO has made a resurgence in the scientific literature and commercial sectors. During this time, studies on using FO for wastewater treatment, seawater desalination, and food processing have been published [4–10]. A recent review [4] gives a comprehensive overview, history, and current state of this technology.

FO processes are based on an osmotic pressure differential across a semipermeable membrane. In FO, the more dilute solution to be concentrated is the feed solution (FS) while the more concentrated solution is referred to as the ‘draw’ solution, brine, or osmotic agent [4]. In this paper, this solution will be referred to as the draw solution (DS).

Unlike pressure-driven membrane processes, the sole driving force for most FO processes is the osmotic pressure differential between the FS and the DS. When compared to pressure-driven membrane processes, such as RO, FO can use significantly less electrical energy. The overall energy cost of FO can be vastly lower than pressure-driven membrane processes if the DS can be easily recovered or discarded using less or lower quality energy. The selection of draw solute, as well as the method of its recovery or disposal, is beyond the scope of this investigation. Previous investigations have employed a variety of osmotically efficient solutes, such as low molecular weight salts (NaCl, MgCl₂, CaCl₂), as surrogates for commercially viable draw solutes [4,9–12].

Membrane selection is one of the most critical aspects to consider in FO process design. Although any selective membrane can be used [4], the most recent studies have found that membranes designed for reverse osmosis do not work well due to extensive mass transfer resistances near the membrane interfaces and within the membrane support structure [5,9,13,14]. These mass transfer resistances, collectively attributed to concentration polarization, require the use of very large osmotic driving forces to sustain adequate water flux. Improvement in membrane design and optimization of the fluid channel hydrodynamics can control the impact of these phenomena, but will not be discussed as part of this study.

Previous investigations of the use of FO in food processing, one application of the work presented here, are focused primarily...
on fruit juice concentration [6–8]. Compared to thermal and pressure-driven membrane technologies, FO processes realize several benefits [15]. Advantages include the low temperature and pressure requirements, which reduce flavor and nutrition degradation, as well as the possibility of treating solutions with high levels of suspended solids. The drawbacks are mainly related to the fact that the FO process is unproven and that DS recovery/disposal may incur energy and/or capital costs. In one particular FO application, these costs have been discussed in greater detail [5].

Further study of the use of FO to process typical food streams is required. For instance, concentration of sugar solutions derived from sugar cane or sugar beet is common practice in many food industries. Concentrated sucrose solutions in particular are widely used in food engineering processes, such as in the formulation of jams and marmalades, bakery products, and candies [16]. Sucrose solutions may also be used (sometimes in combination with salt) to preserve fruits and vegetables [17]. Sucrose solutions also have a role in fermentation processes. For example, in wine making, sugar must be added to grape juice when the sugar content of the grape is not high enough to achieve the desired level of alcohol during fermentation.

These various uses for sugar require a high quality and concentrated sugar source. Sugars that are harvested must be extracted, processed, and concentrated. In the case of sugar crystal formation (granulated sugar), much of the water is removed prior to crystallization. This water is typically removed by evaporation, an energy intensive process [18].

Food processing also tends to generate waste streams containing high concentrations of sugar. For example, juice production from oranges, pineapples, and apples generates large amounts of solid wastes rich in sugar, which, in some cases, could be valorized after the recovery and concentration of the sugar. These solid wastes can be pressed to extract sucrose-rich liquor, but treating this concentrate is the limiting step that increases the final cost of the product. It would, therefore, be desirable to find a way to concentrate or at least to pre-concentrate these sugar-rich solutions to save energy.

The work presented here examines the use of FO as an alternative to other dewatering techniques. Specifically, we investigate the use of FO as a method for sucrose concentration using a commercially available FO membrane and examine the membrane water flux performance as a function of sucrose concentration. Process performance is characterized by determining the maximum degree of concentration in comparison to competing technologies.

### 2. Materials and methods

#### 2.1. Forward osmosis system and module

The experiments were run on a bench-scale laboratory system. A schematic diagram is presented in Fig. 1. The membrane unit was custom built with channels (77 mm long × 26 mm wide × 3 mm deep) on both sides of the membrane. A similar system was described in a previous investigation [19]. Counter current cross-flow was used and the FS and DS were both run in a closed loop. The FS flowed on the active layer of the membrane. In order to increase the mass transport on both sides of the membrane, mesh spacers were inserted within both channels. Variable speed peristaltic pumps (Manostat, Barrington, IL) were used to pump the FS and DS. The temperature of both solutions was controlled to within ±1 °C by a water bath (Neslab, Newington, NH). Water flux (permeate) into the DS was measured by the weight change of the DS reservoir (Denver Instruments, Denver, CO) during a selected period of time.

#### 2.2. Selected membranes

Two membranes were tested in the FO experiments. The selection of these membranes was made by taking into account experimental data from the literature [5,9]. A flat sheet of cellulose membrane (designated CA), provided by Hydration Technologies Inc. (Albany, OR), was used, as it is currently the only commercially available membrane specifically designed for forward osmosis applications. The membrane has been extensively characterized and evaluated for FO desalination processes [4,19]. It has until now not been considered for concentration of sugar solutions. The other membrane chosen for this work was the AG reverse osmosis (RO) membrane from General Electric (Osmonics) as it was used in both our previous investigation on FO desalination [19] and in work published by Dova et al. [9] on direct osmotic concentration of sugar solutions. The AG membrane is a polyamide thin-film composite RO membrane. The work by Dova et al. found the AG membrane to perform the best for direct osmotic concentration of sugar solutions amongst a group of several RO membranes. Specifications of both membranes are listed in Table 1.

#### 2.3. Feed and draw solutions

Different concentrations of sucrose FS were tested in order to simulate various degrees of concentration. Concentrations ranged...
Table 1
Specifications of the commercial FO (CA) and RO (AG) membranes.

<table>
<thead>
<tr>
<th>Material</th>
<th>Salt rejection (%)</th>
<th>Water permeation coefficient (m s(^{-1}) Pa(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>CA Cellulose acetate</td>
<td>95–99 [5]</td>
<td>3.07 (\times) (10^{-12}) [19]</td>
</tr>
<tr>
<td>AG Aromatic polyamide</td>
<td>99.5 [9]</td>
<td>1.19 (\times) (10^{-11}) [9]</td>
</tr>
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</table>

Table 2
Experimental conditions for the FO experiments for sucrose concentration.

<table>
<thead>
<tr>
<th>Sucrose feed solution, M (°Brix)</th>
<th>NaCl draw solution, M</th>
<th>Temperature</th>
</tr>
</thead>
<tbody>
<tr>
<td>CA 0–1.65 (0–48)</td>
<td>2, 4</td>
<td>20\°C, 30\°C</td>
</tr>
<tr>
<td>AG 0–0.7 (0–25)</td>
<td>4</td>
<td>20\°C, 30\°C</td>
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from 0 (deionized water) to 1.65 M sucrose for the FS. The DS was comprised of either 2 or 4 M NaCl. Table 2 lists all combinations of experimental conditions. The crossflow rate was fixed to 1 L min\(^{-1}\) for both solutions, resulting in a crossflow velocity of 21.4 cm/s.

It is well known that NaCl will diffuse across the membrane from the draw solution to the feed solution. This could lead to reduced quality of the concentrated sucrose solutions. However, as reported by McCutcheon et al. [5,19], the salt rejection of the membrane used is relatively high (over 98% for most cases). Next generation FO membranes would likely have much higher salt rejection, comparable to leading RO membranes.

Moreover, in evaluations of pineapple juice concentration by FO, it has been found that (i) the high level of sugar in the product can reduce the saltiness threshold and (ii) the presence of salt has an enhancing effect on sucrose sweetness [11]. Consequently, the effect of NaCl diffusion from the DS to the FS (sucrose) was not considered in this investigation.

2.4. Experimental procedure

A feed stock solution of 2.5 M sucrose was prepared and stored at 4\°C to prevent biological growth. The initial feed solution consisted of deionized water in a 0.5 L feed tank, whereas the initial draw solution was either 2 or 4 M NaCl in a 2 L reservoir. The concentration of sucrose in the feed solution was increased incrementally and permeate flux was gravimetrically measured every minute for each feed solution concentration. When the water flux attained a constant value, a predetermined volume of feed stock solution was added to the feed solution tank to reach the next feed solution concentration. The highest feed solution evaluated was 1.65 M sucrose. The effect of draw solution dilution on the osmotic pressure gradient was not considered since the total permeate volume was not large enough to significantly dilute the relatively large draw solution volume (2L).

3. Results and discussion

3.1. Performance comparison between RO and FO membranes

Fig. 2 shows the permeate flux data for both the CA and AG membranes. Similar to previous investigations, large differences in flux were obtained between the two membranes. The water fluxes achieved for the AG membrane were only 7.7% of the CA membrane fluxes when using deionized feed water and were even worse for tests with FS concentrations up to 0.6 M sucrose. Thus, the CA membrane is clearly more suitable than the AG membrane to concentrate sucrose solution. Similar behavior was observed by McCutcheon et al. [5] when using 0.5 M NaCl feed solutions and a 6 M ammonium–carbon dioxide DS in desalination studies. In that case, the permeate water flux obtained for the AG membrane was around 5% of the CA membrane flux.

In light of the fact that the AG membrane has higher water permeability than the CA membrane (Table 1), the difference in flux performance is due to the membrane support structure as shown in Fig. 3. In these SEM images, the AG membrane contains a significantly thicker support layer (around 160 μm compared to between 30 and 50 μm for the CA membrane). The thick polyester layer comprises the majority of this support layer. The CA membrane, on the other hand, lacks this polyester fabric and instead uses an embedded mesh for structural support. The AG membrane polyester and polysulfone composite support is also hydrophobic.
which would likely prevent full wetting of the support structure, thus producing a reduced water flux when compared to the relatively hydrophilic cellulosic support layer of the CA membrane [20]. The differences in overall support layer thickness, structure, and hydrophilicity critically impact the severity of internal concentration polarization [20], a phenomenon prevalent in anisotropic membranes that is seriously detrimental to FO membrane performance.

### 3.2. Influence of temperature on water flux

Temperature is also shown to have a significant effect on water flux as depicted in Fig. 2. Increasing the temperature from 20 to 30 °C yielded higher water fluxes. Similar behavior was observed in a previous FO study on desalination [19]. The effect of temperature on permeate flux is often described using an Arrhenius-type expression [21]:

$$J_T = J_{T_0} \exp \left( \frac{s}{T} \right)$$

(1)

where $J_T$ is the permeate flux at an arbitrary temperature $T$, $J_{T_0}$ is the permeate flux measured at a reference temperature of 20 °C, and $s$ is an empirical constant that must be evaluated for each membrane. This $s$ value is a function of the activation energy specific to each membrane. Considering the experimental data obtained at both temperatures with the CA membrane, it has been found that the empirical value of $s$ is approximately the same for the test with both the 2 and 4 M DS, giving an average value of 84.32 K.

Using this $s$ value (determined at 20 °C), water flux was fitted at 30 °C for both the 2 and 4 M NaCl draw solutions. Fig. 4 indicates that the predicted water flux matches the experimental data. Thus, the $s$ value obtained at 20 °C could be used to correlate the permeate flux at other operating temperatures. An increased temperature can be used to improve membrane performance (water flux and concentration factor), and this method can be used to predict and optimize operating conditions.

The data presented earlier in Fig. 2 also shows an interesting water flux behavior based on the DS concentration. Using the CA membrane runs at both temperatures, the 4 M DS yields higher flux than the 2 M DS, as would be expected. The relationship between permeate flux and DS concentration, however, is more complicated. As reported in our earlier investigation [19], water flux does not increase proportionally with osmotic driving force due to internal concentration polarization. It is notable that a twofold increase in the DS concentration does not yield a doubling of the permeate flux.

### 3.3. Influence of osmotic pressure driving force on water flux

It is important to analyze the effect of the driving force, in this case the transmembrane osmotic pressure, on the permeate water flux. Fig. 5 presents the dependence of water flux on the osmotic pressure difference between the DS ($\pi_D$) and FS ($\pi_F$) bulk osmotic pressures. Osmotic pressure of the FS was calculated using the Money model [22] for sucrose concentrations up to 0.6 M. Osmotic pressures for higher sucrose concentrations and for the NaCl DS were calculated using a software package from OLI Systems, Inc. (Morris Plains, NJ).

As shown in Fig. 5, water fluxes for the AG membrane were very low. This result was, however, not entirely unexpected. Previous investigations by Dova et al. [9,10] have considered the use of the AG membrane and similar RO membranes for concentration of sugar solutions and, although these membranes performed superbly for pressure-driven flow, they performed poorly for FO applications. In this case, bulk osmotic pressure differences as high as 200 bar led to fluxes lower than 3 L m$^{-2}$ h$^{-1}$.

It is also observed in Fig. 5 that for the same osmotic pressure difference, the flux obtained is higher at 30 °C due to the increase in the water permeability coefficient induced by the lower water viscosity [21]. Increased temperature also results in an increased diffusion coefficient of NaCl and sucrose, mitigating in part the severity of the internal and external concentration polarization phenomena. In addition, it is interesting to note that Fig. 5 indicates that higher transmembrane bulk osmotic pressure differences generated by the 4 M draw solution yield similar water fluxes as those driven by the 2 M draw solution. These differences are due to the more severe internal concentration polarization phenomena associated with more concentrated draw solutions.

Similar results were recounted in previous investigations [4,5,14,19] as analysis concluded that the effective driving force is not the difference between the osmotic pressures of the bulk FS and DS. The effective transmembrane osmotic pressure is determined by the difference in osmotic pressures at the interfaces of the membrane active layer. Ions from the bulk draw solution must diffuse through the membrane support layer to the interior inter-
to increase the concentration factor in RO is to use more energy and stronger pumps, but energy costs, combined with limitations on pump size and the strength of the membrane housings, prohibit this strategy. FO systems have no such limitations as no hydraulic pressure is used. Table 3 shows the concentration factor results and compares the performance of FO and RO. Using the NaCl surrogat... more than twofold the concentration factor reached by RO.

4. Concluding remarks

This study demonstrates that forward osmosis is a viable alternative membrane process for concentration of sucrose solutions. The results demonstrate that, in forward osmosis applications, reverse osmosis membranes perform poorly when compared to membranes designed for forward osmosis. This behavior is mainly attributable to internal concentration polarization. Temperature was found to have a significant effect on the permeate water flux as increased temperature leads to an increase in the draw and feed solute diffusion coefficient and a decrease in water viscosity. It is also shown that water flux does not increase proportionally with the bulk osmotic pressure difference due to the presence of dilutive internal concentration polarization. Minimizing the concentration of the draw solute for a desired concentration factor is preferable as the severity of internal concentration polarization is reduced. However, even with this limitation caused by concentration polarization, concentration factors in excess of 5 were possible with forward osmosis, compared to reported values of, at most, 2.5 with reverse osmosis. Improved performance will be possible with the development of membranes specifically tailored for forward osmosis processes.

Membrane technologies may offer energy efficient methods of concentrating sucrose solutions or other liquid foods when compared to energy intensive evaporative methods. Within the realm of these technologies, forward osmosis may be a more suitable, promising, and sustainable alternative.

Acknowledgments

E. Garcia-Castello gratefully acknowledges the Vicerrectorado de Investigación, Desarrollo e Innovación of the Polytechnic University of Valencia, Spain for the grant Estancia de PDI de la UPV en Centros de Investigación de Prestigio included in the program Apoyo a la Investigación y Desarrollo 2006.

References


Table 3
Sucrose concentration factors for forward osmosis at two different draw solution concentrations obtained in this investigation compared to published data with reverse osmosis [23]. The initial sucrose concentration in the FO experiments was 0.29 M (10°Brix) and the temperature was fixed at 30°C.

<table>
<thead>
<tr>
<th></th>
<th>4 M draw</th>
<th>2 M draw</th>
<th>Reverse osmosis [23]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Final concentration, M (°Brix)</td>
<td>1.65 (56.4)</td>
<td>1.65 (56.4)</td>
<td>0.73 (25)</td>
</tr>
<tr>
<td>Flux (Lm⁻²h⁻¹)</td>
<td>5.84</td>
<td>2.78</td>
<td>11.93</td>
</tr>
<tr>
<td>Concentration factor</td>
<td>5.7</td>
<td>5.7</td>
<td>2.5</td>
</tr>
</tbody>
</table>

Fig. 6. Water flux for the CA membranes as a function of the osmotic pressure difference between the draw and feed solutions normalized by the feed solution osmotic pressure. Data are shown for the runs at both 20 and 30°C. π₀ and πᵣ are the osmotic pressures of the draw and feed solutions, respectively. Experimental conditions: feed solution (sucrose) concentration of 0–165 M (0–48° Brix), draw solution (DS) concentration of 2 or 4 M NaCl, and crossflow rate for draw and feed solutions of 1 Lmin⁻¹ (1.67 × 10⁻³ m³s⁻¹).

The influence of dilutive internal concentration polarization on water flux is represented by considering the relationship between water flux and the transmembrane bulk osmotic pressure difference normalized by the FS osmotic pressure [19]. This data, plotted in Fig. 6, has a logarithmic shape indicating that at low normalized driving forces, water flux strongly increases with increasing osmotic driving force. However, at higher normalized driving force, the increase in water flux diminishes substantially due to increasing severity of internal concentration polarization. It is therefore desirable to use the most dilute DS possible in order to obtain the required concentration factor of the sucrose solution.

3.4. Sucrose concentration factor: comparison to reverse osmosis

Concentration factor is a critical performance criterion in sucrose or other liquid food concentration processes. Traditional membrane processes, like reverse osmosis (RO), have been evaluated [23–26] for concentration of various sugar solutions. RO processes have been found to be effective in concentrating sucrose solutions up to 20–25° Brix from sucrose solutions of 10° Brix (a concentration factor of about 2–2.5). RO was also found to have a primary energy consumption of less than one-seventh of the evaporative method for the same concentration factor [25]. Forward osmosis, however, can use far greater driving forces than those obtained in RO systems while using even less energy. The only way


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