



Sustainable dewatering of grapefruit juice through forward osmosis: Improving membrane performance, fouling control, and product quality



David Inhyuk Kim^{a,b}, Gimun Gwak^c, Min Zhan^a, Seungkwon Hong^{a,*}

^a School of Civil, Environmental & Architectural Engineering, Korea University, 145, Anam-ro, Seongbuk-Gu, Seoul 02841, Republic of Korea

^b Centre for Technology in Water and Wastewater, School of Civil and Environmental Engineering, University of Technology, Sydney (UTS), Broadway, NSW 2007, Australia

^c Center for Water Resource Cycle Research, Korea Institute of Science and Technology, 5, Hwarang-Ro 14-Gil, Seongbuk-Gu, Republic of Korea

ARTICLE INFO

Keywords:

Forward osmosis
Osmotic dewatering
Fouling control
Reverse solute flux
Pressure assisted osmosis

ABSTRACT

Highly enriched grapefruit juice is expected to be obtained through forward osmosis (FO) without degradation of its nutrients. However, this technology is facing several key issues that must be explored to validate the suitability of FO as a dewatering process, namely, membrane performance testing, fouling control, and product quality assessment. In this work, grapefruit juice was dewatered using a commercial thin-film composite FO membrane that exhibited stable performance. The simulation results also suggested that the dewatering could be further enhanced by improving the S value of the current TFC FO membrane. Severe membrane fouling was observed, and it was predominantly due to suspended particles larger than 0.45 μm , such as pectin. However, sustainable osmotic dewatering operation could be attained by implementing appropriate fouling control strategies, such as separating large-sized particles by sedimentation or centrifugation prior to osmosis and recovering the declined water flux by physical cleaning. The dehydrated feed exhibited no significant loss of nutritional value, suggesting that the FO membrane dewatered the juice effectively while retaining its constituents. In addition, the FO process could be further improved to obtain enhanced-quality grapefruit juice by applying pressure to the feed stream or employing a sugar-based draw solution such as glucose.

1. Introduction

Juices extracted or pressed from fruits or vegetables are highly nutritious, as they contain vitamins, minerals, and other beneficial constituents [1]. The dewatering of juice products is necessary to ensure their stability and reduce their preservation and transportation costs [2]. However, the thermal processes commonly applied for concentrating juice products, such as evaporation, have been reported to have detrimental impacts on the sensory and nutritional values of the products. Thus, this conventional technology should be replaced by processes without negative impacts on the sensory or nutritional values of juice products [3–5].

During the past decades, forward osmosis (FO), in which water permeates through a membrane driven by the osmotic pressure difference, has been acknowledged as a potential alternative to the thermal dewatering processes [6]. Highly dewatered juice products are expected to be obtained via FO without nutrient degradation. The use of FO for several types of juices (Table 1) to assess the potential of osmotic dehydration has been investigated in previous studies. The juice products

were found to be highly dehydrated through FO, and no significant loss of nutrients was observed. However, the efficiency of osmotic dewatering was limited by its poor performance, mainly due to the low water permeability of FO membranes. Nevertheless, the recent development of thin-film composite (TFC) FO membranes with high water flux has brought progress in the investigation of FO for juice dewatering [7–9]. The newly developed TFC osmotic membranes are expected to shorten the processing time for juice dewatering compared to the low performance cellulose triacetate (CTA) membranes, potentially resulting in the practical application of FO.

Previous studies on the dewatering performance of FO have focused mainly on water flux and concentration rate. However, several additional issues still need to be examined to verify whether FO is suitable for practical use in the juice industry. Although the newly developed membranes demonstrate significantly improved performance, they have not yet been verified systematically in the context of juice dewatering. Membrane fouling caused by the particulate constituents present in the juices is suspected to rapidly lower the FO efficiency [10–12,14,15]. However, there is a lack of practical work focusing on

* Corresponding author.

E-mail address: skhong21@korea.ac.kr (S. Hong).

<https://doi.org/10.1016/j.memsci.2019.02.031>

Received 11 September 2018; Received in revised form 25 December 2018; Accepted 12 February 2019

Available online 14 February 2019

0376-7388/ © 2019 Elsevier B.V. All rights reserved.

Table 1
Previously reported performance of FO in juice dewatering.

| Juice product | Membrane type | Initial water flux ($L\ m^{-2}\ h^{-1}$) | NaCl draw concentration (M) | Concentration rate (°Brix) | References |
|---------------|---------------|--|-----------------------------|----------------------------|------------|
| Grape | CTA | 8.5 | 6.0 | 4.4 to 54 (12.3 fold) | [10] |
| Beetroot | CTA | 12.4 | 6.0 | 2.3 to 52.0 (22.6 fold) | [10] |
| Pineapple | CTA | 10.5 | 6.0 | 4.4 to 54.0 (12.3 fold) | [10] |
| Orange | CTA | 16.8 | 4.0 | 8.0 to 10.6 (1.3 fold) | [11] |
| Tomato | TFC-RO | 7.1 | 4.0 | 4.3 to 7.5 (1.8 fold) | [12] |
| Sucrose | CTA | 5.8 | 4.0 | 10.0 to 56.4 (5.6 fold) | [13] |

fouling control strategies for sustainable juice dewatering. Additionally, reverse salt diffusion from the draw solutes has raised food safety concerns due to its potential risk to the juice quality. Nonetheless, the effect of reversely diffused salts on the nutritional value of juices has not yet been discussed in detail.

In this study, we systematically investigated the challenges of FO as a dewatering process and their possible improvements to achieve practical dewatering of grapefruit juice. The performance of FO in juice dewatering (i.e., the permeate water and reverse salt fluxes) was first examined using a TFC FO membrane to determine whether current commercial membranes are suitable for application in practical juice dewatering. In addition, a simulation was performed to discover how much further the FO membranes could be improved to obtain a better dewatering rate. Subsequently, the sustainability of FO as a dewatering process was considered through an investigation of the fouling behavior by grapefruit juice. Following the fouling experiments, the fouling reversibility was assessed using several fouling control methods. Lastly, the characteristics of grapefruit juice processed at various concentration rates were analyzed to ensure the safety and quality of the product. We also suggested improvements by which the reverse solute flux could be reduced to enhance the quality of the final product. These results are expected to demonstrate the feasibility of osmotic dewatering for practical use, which has not been reported in the literature.

2. Experimental

2.1. Osmotic membrane and grapefruit juice as feed

A thin-film composite (TFC) membrane (Porifera, Inc., CA) was used throughout the FO experiments in our study. This commercial membrane has shown good stability and performance, exhibiting high water and low salt permeabilities [16,17]. The membrane was received as a flat sheet and stored in deionized (DI) water at 4 °C, followed by rinsing with DI water several times. All the FO tests were carried out in the active layer facing feed solution (AL-FS) membrane orientation.

100% pure Florida grapefruit juice (Natalie's Orchid Island Juice Company, FL) was used as the feed solution to simulate a real juice extraction process. Natural juices that do not undergo a dewatering process such as evaporation have been reported to contain all the nutrients, such as vitamins and minerals, that are found in raw grapefruit, whereas the nutrient content is usually reduced in thermally processed juices [18,19]. The juice feed solution was frozen at −4 °C to ensure its microbiological safety, thawed at 4 °C, and used within 7 days.

2.2. FO experimental setup

The FO tests were performed using a laboratory-scale cross-flow unit as described in our previous studies [20,21]. A custom-built acrylic cell in which both the feed and draw channel had dimensions of 7.7 cm in length, 2.6 cm in width, and 0.3 cm in height provided an effective membrane area of 20.02 cm². The flow rate of both the feed and draw streams was maintained at 0.5 L min^{−1} via recirculation with two variable-speed gear pumps (Cole-Parmer, IL). The temperature of both solutions was adjusted to 20.0 ± 1.0 °C using a thermostat. A reservoir of the bulk draw solution was placed on a digital balance linked to a

computer that collected weight data once per minute. The permeate water flux, J_w , passing through the osmotic membrane was determined based on the change in the draw water weight, whereas the change in the feed water conductivity was measured using a conductivity meter (Hach Company, CO) to calculate the reverse salt flux, J_s .

2.3. FO performance and fouling experiments

The protocol for the performance tests and fouling runs of FO included the following steps. Firstly, a new TFC membrane coupon was placed in the test cell. The inserted membrane was then stabilized in AL-FS mode using DI as both the feed and draw solutions for 20 min. After stabilization, 1 L of grapefruit juice and 2 L of the NaCl draw solution replaced the DI water as the feed and draw, respectively. Different NaCl draw concentrations (1, 2, and 3 M) were employed for the performance tests, whereas 2 M NaCl was used during the fouling runs. Baseline experiments were also carried out to probe the influence of the dilution of the draw solution on the decline in the permeate flux using a synthetic feed mimicking the ionic composition of raw grapefruit juice but containing no foulants. Each test was repeated at least twice by loading different membrane coupons.

2.4. Sedimentation and centrifugation for fouling mitigation

In this study, sedimentation or centrifugation, both of which have been widely used in the food industry, was employed as a pretreatment step to assess their effect on fouling mitigation. Gravitational sedimentation was performed by allowing the particulate matter to settle for 4 h after which only the top of the bulk solution was collected, leaving the settled particulates behind.

Centrifugation was carried out to separate the particulate matter from the bulk grapefruit juice solution. The bulk juice was centrifuged at 3000 × g (4 °C) for 10 min using an Allegra X-15R (Beckman Coulter, CA) to remove particles such as suspended pectin. Component analysis results (not shown in this paper) confirmed that the grapefruit juice was not degraded after centrifugation under these conditions.

2.5. Pressure assisted osmosis (PAO) system and glucose as the draw solution for improved grapefruit juice quality

Two different methods were employed to alleviate the impact of reverse salt flux on the final product quality: the use of a PAO system and the adoption of glucose as the draw solute. During the PAO experiments, the gear pump for the recirculation of the feed solution was replaced by a high-pressure pump (Hydracell, MN). The hydraulic pressure at the feed was applied using a back-pressure regulator positioned at the outlet of the feed channel of the test system. Fine mesh feed spacers were inserted in the draw channel to ensure the mechanical stability of the osmotic membrane [22,23]. The PAO tests were carried out after stabilization at a hydraulic pressure of 6 bar.

In our work, glucose (Sigma-Aldrich, MO), which has been studied as a possible draw solute, was employed in the FO tests as a representative monosaccharide draw solute [24]. The osmotic pressure of this sugar-based draw solution ranged from 34.2 to 58.6 bar for 1.0–2.0 M solutions [24]. The reverse-diffused glucose can be expected

to act as a sweetener in the final juice products. The reverse sugar flux was determined by analyzing the change in the total organic carbon (TOC) with a TOC-V CPH analyzer (Shimadzu, Japan).

2.6. Characterization of the raw and concentrated grapefruit juices

The concentrated grapefruit juices were sampled after the FO tests and subsequently characterized to determine their quality. Cations in the juices were quantified using inductively coupled plasma (ICP) spectroscopy, whereas the concentrations of protein, sucrose, and vitamin C were determined using high-performance liquid chromatography. Lastly, sugar content was analyzed using a sugar refractometer.

3. Results and discussion

3.1. Basic performance evaluation of osmotic dewatering of grapefruit juice

3.1.1. Performance of the TFC-FO membrane for grapefruit juice concentration

The basic performance parameters of FO, including the water and reverse solute fluxes, were first tested using different concentrations of the NaCl draw solution. Fig. 1 presents the initial permeate water and reverse salt fluxes when 1, 2, and 3 M NaCl were used as the draw solution. The initial water flux reached 13.2 and 18.4 L m⁻² h⁻¹ using 2 and 3 M NaCl as the draw solution, respectively. This represents a significantly improved juice dewatering performance compared to results in the literature for cellulose triacetate (CTA) and TFC reverse osmosis (TFC-RO) membranes.

The dewatering rates found in the literature were below 10 L m⁻² h⁻¹ despite the high concentration of the employed draw solutions (4–6 M NaCl), showing very poor performances although high concentrations of the juices were attained. Tomato juice was hardly concentrated, with a water flux of 7.1 L m⁻² h⁻¹ [12], and beetroot juice was dehydrated with a dewatering rate of 12.4 L m⁻² h⁻¹ using 4 and 6 M NaCl draw solutions, respectively [10]. Furthermore, pineapple and grape juices were concentrated 12- and 12.3-fold with initial permeation fluxes of 10.5 and 8.5 L m⁻² h⁻¹, respectively, using a 6 M NaCl draw solution [10]. The satisfactory performance of the tested membrane suggested that the processing time required to effectively concentrate the raw juices could be shortened using high-performance membranes. However, it has to be noted that the processing time is not solely determined only by the membrane performance. Other operating issues, such as the viscosity of the concentrated juice and membrane fouling caused by impurities, should also be considered, since these factors severely decrease the dewatering rate.

3.1.2. Effect of the membrane properties on the water flux in the osmotic dewatering of grapefruit juice

The above analysis demonstrated that the dewatering rate during grapefruit juice concentration could be significantly improved by the progress made in the development of high-performance membrane. We carried out a theoretical sensitivity analysis in which the intrinsic characteristics of the membranes, such as the water permeability, A, the salt permeability, B, and the structural parameter, S, were varied to determine whether the processing performance could be improved further. The improved permeation flux, J_w, obtained by changing the values of A, B, and S can be defined as in Eq. (1) [25]:

$$J_w = A \left(\frac{\pi_{D,b} \exp\left(-\frac{J_w S}{D_d}\right) - \pi_{F,b} \exp\left(\frac{J_w}{k_f}\right)}{1 + \frac{B}{J_w} \left[\exp\left(\frac{J_w}{k_f}\right) - \exp\left(-\frac{J_w S}{D_d}\right) \right]} \right) \quad (1)$$

where $\pi_{D,b}$ and $\pi_{F,b}$ are the osmotic pressures of the bulk draw and feed solutions, respectively, k_f is the mass transfer coefficient of the feed stream, and D_d is the diffusion coefficient of the draw water.

The effect of cross-flow on the mass transfer coefficient can be described as follows (Eqs. (2)–(4) [25]):

$$k_f = \frac{S_h D_f}{d_h} \quad (2)$$

$$Sh = 1.85 \left(Re Sc \frac{d_h}{L} \right)^{0.33} \quad (\text{Laminar flow}) \quad (3)$$

$$Sh = 0.04 Re^{0.75} Sc^{0.33} \quad (\text{Turbulent flow}) \quad (4)$$

where k_f is the solute mass transfer coefficient of the feed water, S_h is the Sherwood number, D_f is the diffusion coefficient of the feed water, and d_h is the hydraulic diameter of the cell channels. S_h is determined using either the laminar or turbulent flow correlation for a rectangular channel, as given above. In these expressions, Re is the Reynolds number, Sc is the Schmidt number, and L is the length of the channel.

The intrinsic parameters of the membrane were characterized using the FO method as described in the literature [26]. The FO tests were run in four stages using DI water as the feed and draw solutions of different concentrations (0.5, 1.0, 1.5, and 2.0 M NaCl). The water and reverse salt fluxes were measured at each stage. The membrane parameters were then estimated by means of the least-square method to minimize the errors between the calculated and experimental fluxes. The derived values of the membrane characteristics, A, B, and S were 2.22 L m⁻² h⁻¹ bar⁻¹, 0.49 L m⁻² h⁻¹, and 269 μm, respectively. In the modeling data, the increase in B was assumed to be proportional to the increase in A due to the trade-off between the water and salt

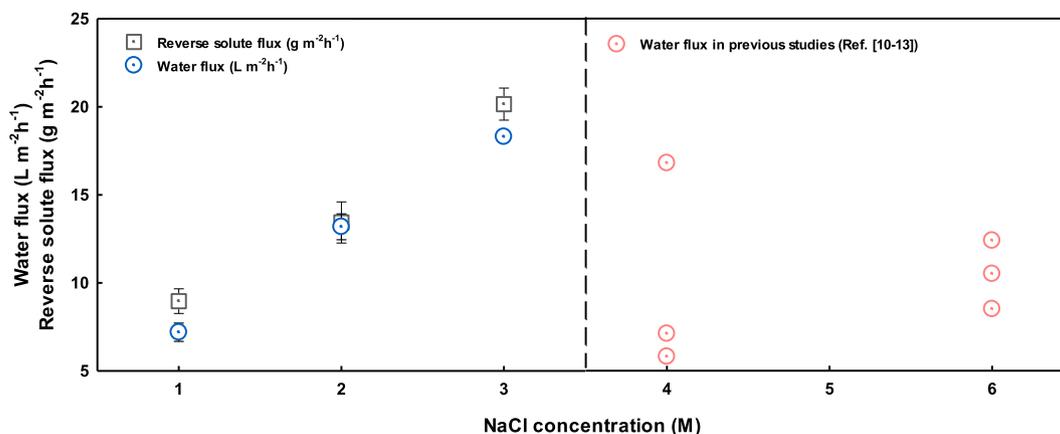


Fig. 1. Initial water and reverse solute fluxes in the dewatering of grapefruit juice using 1–3 M NaCl as the draw solution, and comparison with previously reported results. The cross-flow and temperature during the experiments were fixed to 10.7 cm s⁻¹ and 20.0 ± 1.0 °C, respectively. The average J_s/J_w throughout the tests was 1.14 g L⁻¹. The water and reverse solute fluxes were determined for the first 5 min.

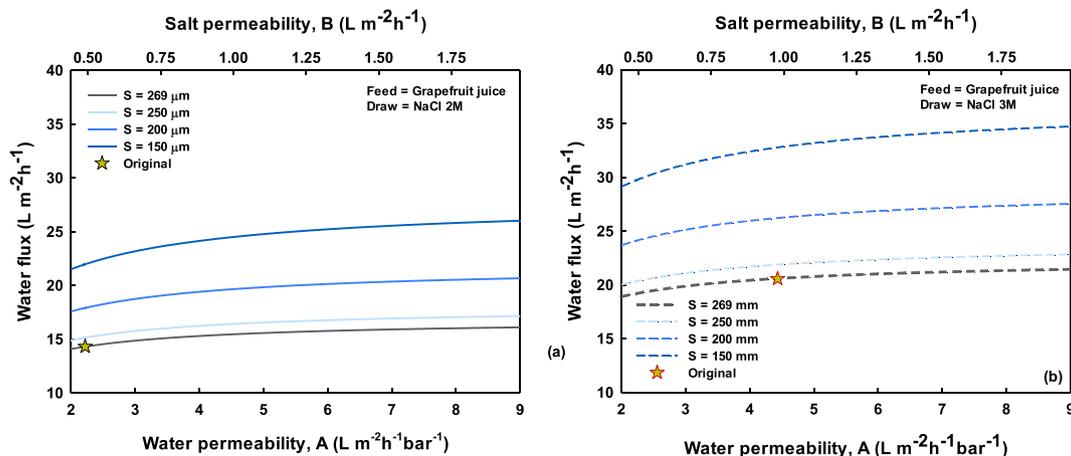


Fig. 2. Effects of the membrane characteristics (A, B, and S) on the dewatering rate in FO using (a) 2 M and (b) 3 M NaCl as the draw solution. Modeling conditions: cross-flow velocity: 10.7 cm s^{-1} , diffusion coefficient: $1.23 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$, dynamic viscosity: 0.98 cP . The lines represent the simulation results, and the star symbols indicate the model fluxes based on the experimentally determined membrane characteristics.

permeabilities [27]. The osmotic pressure of the bulk grapefruit juice feed solution was measured using an Osmomat 030 (Gonotec GmbH, Germany).

As shown in Fig. 2(a), the water flux increased significantly with an increase in S, whereas increasing the value of A had only a minor impact on the dewatering performance. Specifically, the water flux increased from 14.3 to $21.9 \text{ L m}^{-2} \text{ h}^{-1}$ (a 53.1% increase) when S was reduced from 269 to $150 \mu\text{m}$. In contrast, the water flux increased slightly from 14.3 to $16.1 \text{ L m}^{-2} \text{ h}^{-1}$ (a 12.6% increase) when A was increased by a factor of 4.05 (from 2.22 to $9.00 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$). This was likely because the water permeability of the current TFC FO membrane was sufficiently high, and the increase in the water flux with further increase in A was limited due to the enhanced external concentration polarization (ECP) and the dilutive internal concentration polarization (ICP) [28]. Improving the water flux by reducing the structural parameter, which is inversely proportional to the intensity of the dilutive ICP is, therefore, more effective to overcome this challenging issue. The increasing trend in the water flux became more evident as the S value was decreased while operating with a 3 M NaCl draw solution (Fig. 2(b)). These results suggest that the osmotic dewatering performance of grapefruit juice could be further enhanced by developing improved osmotic membranes. In particular, the processing time could be shortened to meet the requirements for effective osmotic dewatering for the concentration of grapefruit juice by developing a support layer with a low structural parameter. However, it has to be noted that the particulate constituents in grapefruit juice can severely risk the dewatering performance by fouling the osmotic membrane. Therefore, investigating the membrane fouling behavior and implementing proper fouling control strategies is also critical.

3.2. Mitigation of fouling in grapefruit juice dehydration

3.2.1. Fouling behavior in FO during grapefruit juice processing

Previous studies of osmotic dewatering have barely examined the impact of the fouling caused by particle accumulation on FO performance, and have discussed only the recovery of the flux decline by simple physical cleaning [12]. However, as grapefruit juice contains suspended pulps such as pectin, serious concerns arise regarding the significant decrease in the dewatering rate due to the instant accumulation of pulp on the membrane active layer surface. In addition, detachment of the pulp is rather challenging. Thus, in this study, the flux decline behavior during the concentration of grapefruit juice was investigated by performing several fouling tests.

The results of the fouling runs using non-filtered grapefruit juice and juice passed through 0.45 and $1 \mu\text{m}$ filters are shown in Fig. 3. For the

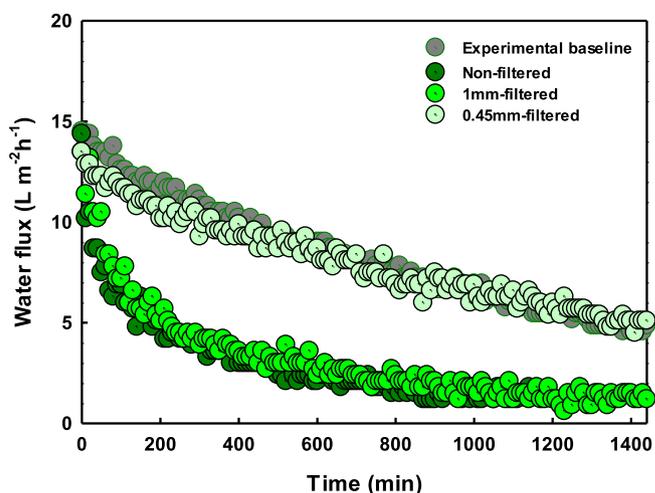


Fig. 3. Flux decline caused by grapefruit juices using three filtration approaches (no filter, $1 \mu\text{m}$ filter, and $0.45 \mu\text{m}$ filter). During the fouling tests, 2 M NaCl was employed as the draw solution, and the temperature and cross-flow velocity were adjusted to $20 \text{ }^\circ\text{C}$ and 10.7 cm s^{-1} , respectively. The experimental baseline presents the flux decline caused by dilution of the draw solution.

non-filtered and $1 \mu\text{m}$ -filtered grapefruit juice, the water flux declined immediately after the start of the fouling test, whereas very little fouling of the TFC membrane occurred when only substances smaller than $0.45 \mu\text{m}$ were present in the bulk feed solution. These results demonstrated that the fouling layer that severely blocks the osmotic membrane during grapefruit juice concentration comprises mainly large-sized suspended constituents. Furthermore, a previous FO study on orange juice dewatering revealed that calcium ions interact with the polysaccharides found in pectin, consequently forming a gel layer that intensifies the fouling [29]. These results imply that suspended pectin, which is abundant in grapefruit juice, should be properly managed to achieve sustainable grapefruit juice dewatering through FO.

3.2.2. Fouling control in FO grapefruit juice processing

Suitable control of membrane fouling during grapefruit juice dewatering is a critical issue for sustainable operation. However, this membrane fouling cannot be controlled by most existing technologies, such as chemical dosing, separation of constituents by micro-/ultra-filtration, and chemical cleaning, as they may degrade the juice quality or reduce its nutritional content. Therefore, in our work, hydraulic methods were adopted; namely, the cross-flow rate was increased two-

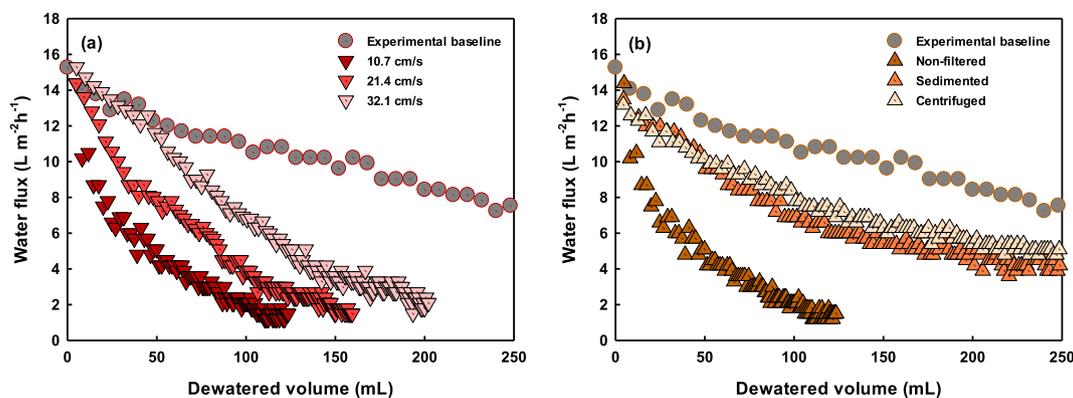


Fig. 4. Impact of (a) hydraulic fouling control (increasing the cross-flow rate) and (b) sedimentation/centrifugation pretreatment on the flux decline during grapefruit juice dehydration. 2 M NaCl was employed as the draw solution. The experimental baseline represents the flux decline due to the dilution of the draw solution.

or three-fold (21.4 and 32.1 cm s⁻¹) relative to the base condition (10.7 cm s⁻¹). The faster cross-flow relieved the flux decline during the early stages of the FO run. However, increasing the shear-force was not effective for longer runs (Fig. 4(a)). These findings are in contrast with a previous work on FO membrane fouling, in which the flux decline was sufficiently diminished when a high cross-flow rate was applied [30]. The mitigation of the fouling in the previous study was attributed to the enhanced shear force generated by the accelerated cross-flow, which hindered the accumulation of dissolved foulants on the membrane surface. The conflicting results in this work likely originated from the size of the fouling matters, which were larger than 0.45 μm. The strong shear force interfered with the adhesion of the large-sized suspended particles on the membrane surface; however, it was not effective at flushing the particles once they were accumulated on the surface, due to their high weight.

In our study, the suspended constituents larger than 0.45 μm were identified as the primary cause of fouling during juice dewatering, and could not be mitigated using the hydraulic method. Therefore, these problematic substances were separated from the bulk feed solution using simple pretreatment methods such as sedimentation and centrifugation. Unlike in micro- or ultra-filtration, the physically separated constituents could be reintroduced to the grapefruit juice solution after dewatering, thus avoiding nutritional losses during the pretreatment stage. As shown in Fig. 4(b), the water flux declined less rapidly after sedimentation/centrifugation, suggesting that these pretreatment strategies were better choices for relieving membrane fouling. This is because the majority of the suspended particles, such as pectin, could be separated by the pretreatment methods. More advanced separation technologies are likely to be more effective in removing the particulate fouling matter. However, sedimentation or centrifugation is a more ideal option, as these methods allow easy reconstitution of the grapefruit juice by re-introducing the extracted constituents to the dewatered feed solution.

3.2.3. Reversibility of membrane fouling during long-term FO juice dewatering operation

Even though membrane fouling can be considerably reduced by centrifugation or sedimentation, the small amount of flux decline has to be managed for sustainable grapefruit juice dewatering via FO. Therefore, the fouling reversibility during long-term operation of FO grapefruit juice dewatering was examined. Physical cleaning by increasing the cross-flow rate three-fold (32.1 cm s⁻¹) relative to the base rate was carried out after each cycle for five cycles. Fig. 5(a) shows the flux decline produced by centrifuged grapefruit juice and the corresponding flux recovery by physical cleaning after each fouling cycle. The decline in the dewatering rate was mostly recovered by simple physical flushing after each cycle, indicating that the fouling matter in

the centrifuged grapefruit juice could be readily detached by the strong shear force [30]. Thus, the dewatering of grapefruit juice was validated as sustainable when regular physical cleaning was applied and suspended pulps such as pectin were pre-separated. This was further confirmed by scanning electron microscopy (SEM) images of the surfaces of the virgin membrane and the physically cleaned membrane after the 5th cycle. No distinct deposits were detected on the membrane surface after long-term operation, as demonstrated by the image in Fig. 5(c), which shows a similar surface morphology to that of the virgin membrane (Fig. 5(b)).

3.3. Characteristics of the concentrated grapefruit juice and improvements to enhance juice quality

3.3.1. Measurement of the quality of the final grapefruit juice product

The quality of concentrated product is regarded as the most crucial issue in FO dewatering, since the reverse salt flux from the draw solution is suspected to degrade the quality of FO-processed juice products. Therefore, the accumulation of diffused salts in the feed after FO operation and their impact on the quality of the juice should be investigated. Most previous studies on the use of FO for juice dewatering evaluated only the concentration factor by measuring the sugar content of the final products [11,13]; only one study measured the amount of diffused salts in the final product [31]. Hence, more careful discussion is required to assure the quality of juice products concentrated using a TFC membrane.

The major constituents of 10, 30, 50, and 75% concentrated grapefruit juices are listed in Table 2. The results showed that the contents of each component remained the same within the allowable margin of error, indicating that all the measured components of grapefruit juice were retained without any significant loss or degradation. This demonstrates that FO can successfully be used to obtain dehydrated juices with high product quality. Nevertheless, the increased sodium content in the processed juice due to reverse salt diffusion raises concerns about unfavorable effects on sensory values.

For a more complete understanding, the dewatered juices were reconstituted to the same sugar content as the raw grapefruit juice. The sodium concentrations of the reconstituted solutions are shown in Fig. 6. As shown, the sodium content of the reconstituted juices increased linearly with the concentration rate of the juices, reaching 18.1 mg/100 mL (i.e., 0.0181% (w/v)) when the grapefruit juice was concentrated four-fold and re-diluted. However, such a low salt content could enhance the sweetness of the processed grapefruit juice rather than deteriorating its taste and odor [32,33]. The amount of reversely diffused salt was very small in comparison with that reported in a previous study on the dewatering of pineapple juice, in which the salt concentration of the processed juice reached a peak amount of 0.58%

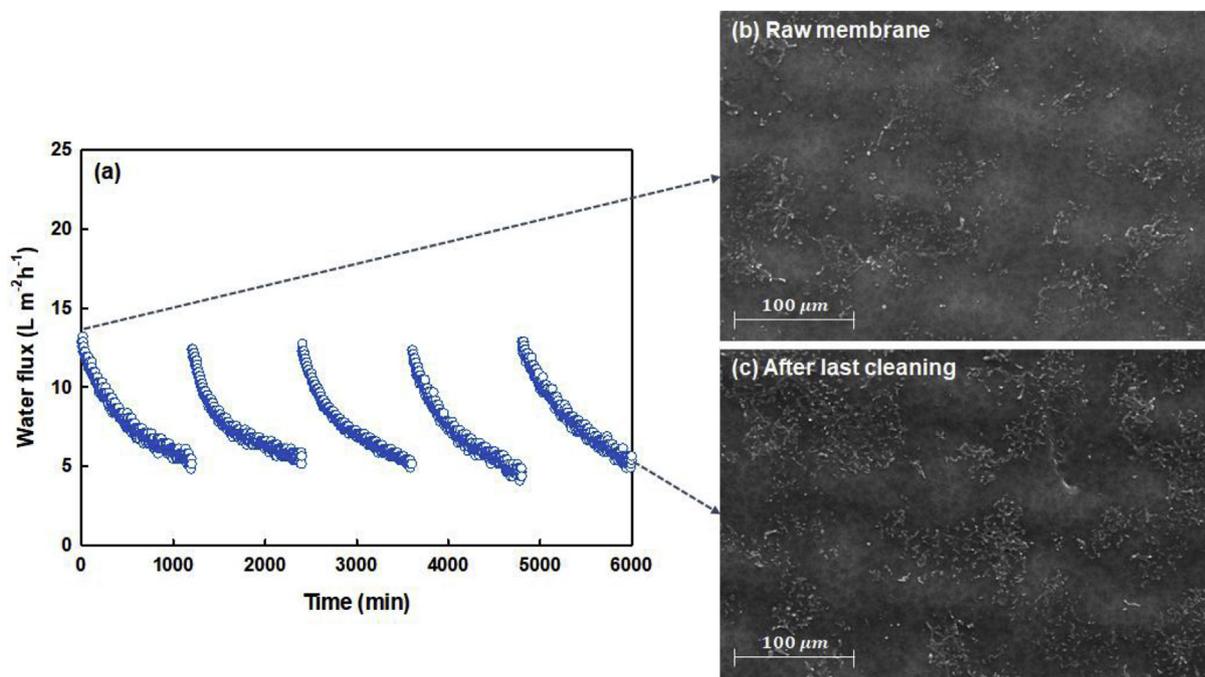


Fig. 5. Long-term grapefruit juice dewatering operation using FO. Physical cleaning was applied after each 20 h cycle by increasing the cross-flow rate to 32.1 cm s⁻¹. (a) Flux decline curve as a function of time during FO. SEM images of (b) the virgin membrane and (c) the cleaned membrane after the 5th cycle. The centrifuged grapefruit juice feed and the 2 M NaCl draw solution were replaced at the initiation of each fouling cycle. The temperature and cross-flow velocity were adjusted to 20 °C and 10.7 cm s⁻¹, respectively.

(37.9 times higher than in our study) [10].

3.3.2. Improvement of the product quality using pressure assisted osmosis and glucose as the draw solute

The small amount of reversely diffused salts present after osmotic dewatering might raise concerns regarding juice quality. To improve the quality of the grapefruit juice and to ensure the safety of the dewatered solution, proper strategies to control salt diffusion should be applied. In this work, PAO, which has been studied for the enhancement of water flux in wastewater treatment or seawater desalination, was introduced because of its potential to reduce reverse salt diffusion in dewatering processes [28,34]. A sugar-based draw solution, glucose, was also tested, as reversely diffused glucose could also act as an additive to enhance or preserve the sweetness of the grapefruit juice.

The water and specific reverse salt fluxes during PAO with applied pressures in the 0–6 bar range are shown in Fig. 7(a). As shown, this pressure applied system did not significantly improve the dewatering rate (13.2–14.8 L m⁻² h⁻¹ at applied pressures of 0–6 bar). Meanwhile, a dramatic reduction of J_s/J_w (from 1.02 to 0.37 g L⁻¹, a 64.1% decrease) was observed using the PAO process. This can be attributed to the great reduction in the effective concentration difference of the salts

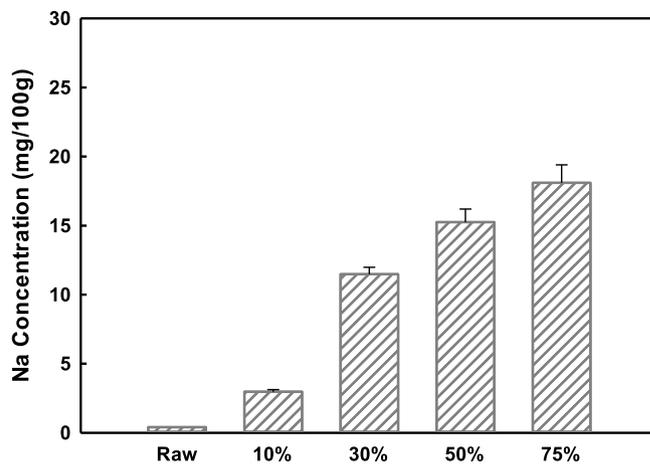


Fig. 6. Na concentration of the raw and reconstituted grapefruit juices. The sugar content of each processed juice was adjusted to 11.4 °Brix by adding DI water.

Table 2
Characteristics of raw and 10, 30, 50, and 75% concentrated grapefruit juices.

| Constituent | Unit | Raw | Concentrated | | | |
|---------------|------------|-------------|--------------|-------------|-------------|-------------|
| | | | 10% | 30% | 50% | 75% |
| Protein | (g/100 g) | 0.4 ± 0.0 | 0.4 ± 0.0 | 0.6 ± 0.1 | 0.8 ± 0.0 | 1.7 ± 0.1 |
| Sucrose | (g/100 g) | 2.5 ± 0.0 | 2.7 ± 0.0 | 3.5 ± 0.1 | 5.1 ± 0.0 | 10.2 ± 0.1 |
| Vitamin C | (mg/100 g) | 17.4 ± 0.2 | 19.0 ± 0.8 | 24.9 ± 0.5 | 35.0 ± 0.8 | 70.2 ± 1.0 |
| Sugar content | (° Brix) | 11.4 ± 0.0 | 12.4 ± 0.0 | 16.5 ± 0.1 | 22.2 ± 0.1 | 45.1 ± 0.1 |
| Ca | (mg/100 g) | 6.3 ± 0.1 | 7.3 ± 0.3 | 9.1 ± 0.1 | 12.8 ± 0.2 | 25.4 ± 0.3 |
| K | (mg/100 g) | 102.5 ± 0.2 | 114.6 ± 1.0 | 146.7 ± 1.4 | 205.5 ± 0.4 | 410.8 ± 1.2 |
| Mg | (mg/100 g) | 8.6 ± 0.1 | 9.9 ± 0.0 | 12.5 ± 0.5 | 17.5 ± 0.5 | 35.9 ± 0.5 |
| Na | (mg/100 g) | 0.4 ± 0.0 | 3.3 ± 0.3 | 16.4 ± 1.0 | 30.5 ± 1.9 | 72.4 ± 2.6 |
| P | (mg/100 g) | 9.5 ± 0.0 | 10.5 ± 0.0 | 14.0 ± 0.3 | 18.8 ± 0.2 | 38.3 ± 0.3 |

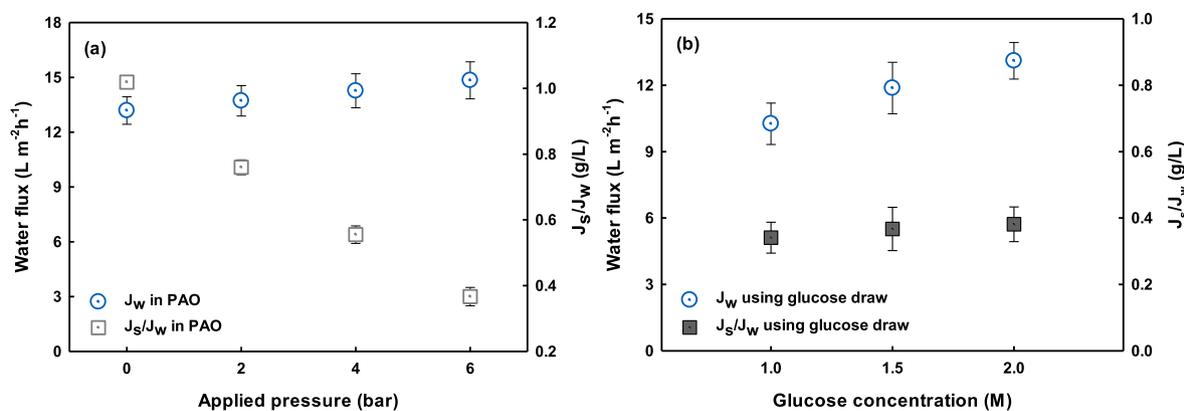


Fig. 7. Initial dewatering rate and specific reverse solute flux in (a) PAO at an applied pressure of 0–6 bar using 2 M NaCl as the draw solute, and (b) FO using 1–2 M of glucose as the draw solute. The fluxes were determined for the first 5 min.

Table 3

Quality of the 50% concentrated final product obtained using FO with glucose as the draw solute and PAO at 6 bar of hydraulic pressure.

| Constituent | Unit | Concentration | | |
|---------------|------------|-----------------------|--------------------------|--------------|
| | | NaCl as draw solution | Glucose as draw solution | PAO at 6 bar |
| Sugar content | (° Brix) | 22.2 ± 0.1 | 22.4 ± 0.1 | 22.2 ± 0.1 |
| Na | (mg/100 g) | 30.5 ± 1.9 | 0.5 ± 0.2 | 11.4 ± 1.1 |

across the membrane due to both the intensified enhanced ECP and dilutive ICP, which were influenced by the hydraulic pressure [35]. PAO effectively limits the reverse diffusion of salts, as the hydraulic-pressure-driven water flux increases the effective feed concentration on the active layer surface while simultaneously diluting the effective draw concentration within the support layer [35]. Fig. 7(b) shows the water flux and the corresponding J_s/J_w at glucose draw concentrations of 1.0–2.0 M. The water flux during the FO dewatering of grapefruit juice was relatively low ($13.1 \text{ L m}^{-2} \text{ h}^{-1}$) even when 2.0 M glucose was employed as the draw solution. However, due to its low osmotic pressure yield and resulting low specific solute flux, glucose could be utilized as a suitable draw solute. Moreover, the small amount of diffused sugar would positively affect the concentrated juice product.

The quality of the 50% dehydrated grapefruit juices processed by FO using a glucose draw and PAO at 6 bar is presented in Table 3. Only the sugar content and sodium concentration were analyzed, since the other constituents in the juice were shown to be retained with no significant loss. The sodium content of the juice concentrate remained very low when glucose was used as the draw solution, whereas the sugar content increased slightly due to reverse diffusion of glucose, demonstrating the effectiveness of the glucose draw solute as an additive in juice dewatering. When PAO was utilized with a pressure of 6 bar, the amount of accumulated salt was greatly reduced as expected (from 30.5 to 11.4 mg/100 g, a 62.6% decrease). The results show the potential of PAO as an osmotic dewatering process to provide improved food quality by significantly lowering salt diffusion.

4. Conclusion

The challenges of applying FO in food processing, as well as possible improvements to the process, have been examined from a practical perspective. Specifically, the performance of FO for the concentration of grapefruit juice was evaluated. In addition, the extent and mechanisms of FO membrane fouling and fouling-mitigation approaches were also investigated. Lastly, the safety and quality of the final product were determined. The major conclusions that have been drawn from the

present work are summarized below:

- Compared to the poor performance reported in previous FO studies, the successful osmotic concentration of grapefruit juice was achieved using a TFC membrane, with a stable dewatering rate and low salt permeability. The dewatering rate could be further improved by reducing the structural parameter value, S , of the osmotic membrane.
- Membrane fouling severely decreased the water permeation rate due to the suspended matter such as pectin in the grapefruit juice. Simple sedimentation and centrifugation noticeably alleviated the water flux decline by isolating the suspended constituents from the bulk juice. Increasing the cross-flow rate was found to be ineffective for fouling mitigation. The decline in the water flux was readily recovered by simple physical cleaning, leading to the sustainable operation of FO dewatering.
- The draw solutes barely diffused into the juice, and thus, high-quality processed juice was obtained. Except for an increase in sodium, the contents of the nutritional substances were maintained after concentration. Applying pressure on the feed side helped to further enhance the quality of the juice product by inhibiting reverse salt diffusion. In addition, glucose can be employed as the draw solution; the reversely diffused glucose acts a sweetener, and thus does not adversely affect the quality of the dewatered feed.

Acknowledgments

This work was supported by Korea Environment Industry & Technology Institute (KEITI) through Plant Research Program, funded by Korea Ministry of Environment (MOE)(1615009988).

References

- [1] D.W. Sun, *Emerging Technologies for Food Processing*, Elsevier, 2014.
- [2] N. Rastogi, *Reverse Osmosis and Forward Osmosis for the Concentration of Fruit Juices*, Elsevier, 2018.
- [3] V. Sant'Anna, L.D.F. Marczak, I.C. Tessaro, Membrane concentration of liquid foods by forward osmosis: process and quality view, *J. Food Eng.* 111 (2012) 483–489.
- [4] B. Jiao, A. Cassano, E. Drioli, Recent advances on membrane processes for the concentration of fruit juices: a review, *J. Food Eng.* 63 (2004) 303–324.
- [5] N.K. Rastogi, Opportunities and challenges in application of forward osmosis in food processing, *Crit. Rev. Food Sci. Nutr.* 56 (2016) 266–291.
- [6] G. Gwak, D.I. Kim, S. Hong, New industrial application of forward osmosis (FO): precious metal recovery from printed circuit board (PCB) plant wastewater, *J. Membr. Sci.* 552 (2018) 234–242.
- [7] N.Y. Yip, A. Tiraferri, W.A. Phillip, J.D. Schiffman, M. Elimelech, High performance thin-film composite forward osmosis membrane, *Environ. Sci. Technol.* 44 (2010) 3812–3818.
- [8] J. Wei, C. Qiu, C.Y. Tang, R. Wang, A.G. Fane, Synthesis and characterization of flat-sheet thin film composite forward osmosis membranes, *J. Membr. Sci.* 372 (2011) 292–302.
- [9] T.S. Chung, S. Zhang, K.Y. Wang, J. Su, M.M. Ling, Forward osmosis processes:

- yesterday, today and tomorrow, *Desalination* 287 (2012) 78–81.
- [10] C.A. Nayak, S.S. Valluri, N.K. Rastogi, Effect of high or low molecular weight of components of feed on transmembrane flux during forward osmosis, *J. Food Eng.* 106 (2011) 48–52.
- [11] E.M. Garcia-Castello, J.R. McCutcheon, Dewatering press liquor derived from orange production by forward osmosis, *J. Membr. Sci.* 372 (2011) 97–101.
- [12] K.B. Petrotos, A.V. Tsiadi, E. Poirazis, D. Papadopoulos, H. Petropakis, P. Gkoutosidis, A description of a flat geometry direct osmotic concentrator to concentrate tomato juice at ambient temperature and low pressure, *J. Food Eng.* 97 (2010) 235–242.
- [13] E.M. Garcia-Castello, J.R. McCutcheon, M. Elimelech, Performance evaluation of sucrose concentration using forward osmosis, *J. Membr. Sci.* 338 (2009) 61–66.
- [14] C.A. Nayak, N.K. Rastogi, Forward osmosis for the concentration of anthocyanin from *Garcinia indica* Choisy, *Separ. Purif. Technol.* 71 (2010) 144–151.
- [15] C.A. Nayak, N.K. Rastogi, Comparison of osmotic membrane distillation and forward osmosis membrane processes for concentration of anthocyanin, *Desalination Water Treat.* 16 (2010) 134–145.
- [16] G. Blandin, H. Vervoort, P. Le-Clech, A.R. Verliefde, Fouling and cleaning of high permeability forward osmosis membranes, *J. Water Process Eng.* 9 (2016) 161–169.
- [17] G. Gwak, S. Hong, New approach for scaling control in forward osmosis (FO) by using an antiscalant-blended draw solution, *J. Membr. Sci.* 530 (2017) 95–103.
- [18] B. Tiwari, C. O'Donnell, P. Cullen, Effect of non thermal processing technologies on the anthocyanin content of fruit juices, *Trends Food Sci. Technol.* 20 (2009) 137–145.
- [19] J. Yi, B.T. Kebede, D.N.H. Dang, C. Buvé, T. Grauwet, A. Van Loey, X. Hu, M. Hendrickx, Quality change during high pressure processing and thermal processing of cloudy apple juice, *LWT-Food Sci. Technol.* 75 (2017) 85–92.
- [20] D.I. Kim, J. Choi, S. Hong, Evaluation on suitability of osmotic dewatering through forward osmosis (FO) for xylose concentration, *Separ. Purif. Technol.* 191 (2018) 225–232.
- [21] R. Gonzales, M. Park, L. Tijing, D. Han, S. Phuntsho, H. Shon, Modification of nanofiber support layer for thin film composite forward osmosis membranes via layer-by-layer polyelectrolyte deposition, *Membranes* 8 (2018) 70.
- [22] B.G. Choi, D.I. Kim, S. Hong, Fouling evaluation and mechanisms in a FO-RO hybrid process for direct potable reuse, *J. Membr. Sci.* 520 (2016) 89–98.
- [23] J. Kim, B. Kim, D.I. Kim, S. Hong, Evaluation of apparent membrane performance parameters in pressure retarded osmosis processes under varying draw pressures and with draw solutions containing organics, *J. Membr. Sci.* 493 (2015) 636–644.
- [24] Q. Long, G. Qi, Y. Wang, Evaluation of renewable gluconate salts as draw solutes in forward osmosis process, *ACS Sustain. Chem. Eng.* 4 (2015) 85–93.
- [25] J.R. McCutcheon, M. Elimelech, Influence of concentrative and dilutive internal concentration polarization on flux behavior in forward osmosis, *J. Membr. Sci.* 284 (2006) 237–247.
- [26] A. Tiraferri, N.Y. Yip, A.P. Straub, S.R.-V. Castrillon, M. Elimelech, A method for the simultaneous determination of transport and structural parameters of forward osmosis membranes, *J. Membr. Sci.* 444 (2013) 523–538.
- [27] N.Y. Yip, M. Elimelech, Performance limiting effects in power generation from salinity gradients by pressure retarded osmosis, *Environ. Sci. Technol.* 45 (2011) 10273–10282.
- [28] J. Kim, D.I. Kim, S. Hong, Analysis of an osmotically-enhanced dewatering process for the treatment of highly saline (waste) waters, *J. Membr. Sci.* 548 (2018) 685–693.
- [29] B. Chanukya, N.K. Rastogi, Ultrasound assisted forward osmosis concentration of fruit juice and natural colorant, *Ultrason. Sonochem.* 34 (2017) 426–435.
- [30] C. Boo, M. Elimelech, S. Hong, Fouling control in a forward osmosis process integrating seawater desalination and wastewater reclamation, *J. Membr. Sci.* 444 (2013) 148–156.
- [31] B.R. Babu, N. Rastogi, K. Raghavarao, Effect of process parameters on transmembrane flux during direct osmosis, *J. Membr. Sci.* 280 (2006) 185–194.
- [32] G. Sacchetti, A. Gianotti, M. Dalla Rosa, Sucrose–salt combined effects on mass transfer kinetics and product acceptability. Study on apple osmotic treatments, *J. Food Eng.* 49 (2001) 163–173.
- [33] B. Ade-Omowaye, N. Rastogi, A. Angersbach, D. Knorr, Osmotic dehydration behavior of red paprika (*Capsicum annum* L.), *J. Food Sci.* 67 (2002) 1790–1796.
- [34] G. Blandin, D.T. Myat, A.R. Verliefde, P. Le-Clech, Pressure assisted osmosis using nanofiltration membranes (PAO-NF): towards higher efficiency osmotic processes, *J. Membr. Sci.* 533 (2017) 250–260.
- [35] Y. Oh, S. Lee, M. Elimelech, S. Lee, S. Hong, Effect of hydraulic pressure and membrane orientation on water flux and reverse solute flux in pressure assisted osmosis, *J. Membr. Sci.* 465 (2014) 159–166.