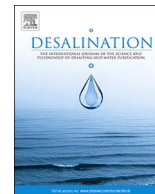




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## Analysis of enhancing water flux and reducing reverse solute flux in pressure assisted forward osmosis process

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## ABSTRACT

Pressure assisted osmosis (PAO) has been recently suggested as a way to overcome the current limitations of forward osmosis (FO), since water flux can be increased by additional hydraulic driving force. To validate its feasibility more fundamentally, the effect of hydraulic pressure combined with osmotically driven FO process was evaluated experimentally, and compared with theoretical modeling. Four different FO and NF membranes were selected and their performance characteristics were determined by both RO- and FO-based methods, since PAO is a simultaneous osmotic- and pressure-driven membrane process. The degree of enhancing water flux and reducing reverse solute flux (RSF) in the PAO process clearly differed according to the membrane type and their performance parameters. Modeling PAO performance, using the values of A, B, and S determined from both RO- and FO-based methods, often failed to exactly match experimental observations, particularly for thin film composite (TFC) FO membranes, suggesting that PAO membrane performance parameters are apparently pressure-dependent. Discernible compaction in the support layer of TFC FO membranes was identified and confirmed by FO tests at different operating modes and SEM analysis, partially explaining large variations in S values under pressurized PAO operation and thus resulting large deviation from theoretical predictions.

### 1. Introduction

The forward osmosis (FO) process has attracted a great attention over last decade as an emerging technology for desalination and water treatment [1–5]. Compared to a conventional reverse osmosis (RO) process, the FO process exhibits great promise in terms of lower energy requirements [6,7] and higher fouling reversibility [8–13]. With these great advantages, the range of possible applications of FO technology is expanded to now include not only food processing [14,15], but also the desalination of high saline water such as shale gas wastewater treatment [16–18].

Despite the enormous potential of FO, several limitations impede its real-world application. One crucial issue still to be resolved is the concentration polarization (CP), which reduces the effective osmotic gradient across the membrane, resulting in lower-than-expected water flux [19–22]. Especially, the internal CP (ICP) which occurs within the support layer of FO membranes decreases the effective driving force significantly more than the external CP (ECP) does. Therefore, the general efficiency of the FO process, including its water flux and productivity, is inevitably lower than that of pressure-driven membrane

technologies (such as RO). Since the CP phenomena is caused by properties inherent to the FO membrane which cannot be eliminated, meaningful advances in the FO process require an in-depth understanding of CP and technical developments mitigating its deleterious effects.

The newly developed concept, pressure assisted osmosis (PAO), may be a desirable solution which overcomes the current limitations of FO [23]. The PAO incorporates both the FO and RO processes, and relies on the application of hydraulic pressure to the feed solution. Water flux is increased by the synergistic effects of the osmotic and hydraulic pressures, thus improving the overall efficiency of the osmosis process [24–26]. Unfortunately, only few papers on this novel idea have been published and the technology is still regarded as immature.

In order to evaluate the feasibility of PAO, several researchers have attempted to examine the effect of hydraulic pressure on system performance. Previous study conducted simple PAO experiments with cellulose triacetate (CTA) and thin-film composite (TFC) membranes, and it reported that the performance of TFC FO membrane was slightly improved by the additional pressure and thus more suitable for PAO process [25]. Other research works have focused on the impact of the

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spacer in the PAO system, since the hydraulic pressure may lead to undesirable membrane deformation or fracture. Blandin et al. [24] demonstrated the significance of employing appropriate spacer to avoid membrane deformation. This phenomenon of membrane deformation by hydraulic pressure has been also reported pressure retarded osmosis (PRO) studies [27,28]. To prevent membrane deformation by the applied pressure, a new support method, using a bundle of permeate carriers and porous stainless steel as feed spacers, was suggested by Oh et al. [26]. Membrane deformation was effectively prevented using the proposed support guide. However, under specific operating conditions, the spacer caused some disruption, such as a shadow effect. Moreover, apparent performance parameter, solute permeability, determined by a new method of characterizing PRO membrane varied significantly with the hydraulic pressure applied to draw solution [20].

As we summarized, most previous studies are limited to simple evaluation of PAO performance improvement without fundamental examination on the synergetic effect of combining osmotic- and pressure-driven membrane processes. Thus, to validate the feasibility of PAO technology, the mechanism of enhancing water flux and reducing reverse solute flux (RSF) by applying additional pressure to feed water in a typical FO process should be understood theoretically and verified with well-produced experimental observations. Thus, this study was designed and conducted systematically to analyze the fundamental mechanisms of improving FO performance by providing additional pressure. First, various FO and NF membranes were evaluated for PAO process by utilizing two different methods to determine their performance characteristics. A series of PAO experiments were then conducted to critically assess the synergetic impact of hydraulic pressure assisting osmotically driven water flux as well as limiting RSF. These experimental observations were further examined with theoretical predictions based on performance parameters determined by RO and FO systems. A series of examinations including FO tests and SEM analyses were also carried out to investigate the impact of hydraulic pressure on the physical structures of the membranes. This work is expected to significantly improve our experimental analysis on PAO process which is characterized to be between pressure driven (e.g. RO) and osmotic driven (e.g. FO) processes.

## 2. Materials and methods

### 2.1. PAO membranes

Four different commercial flat-sheet osmosis membranes were selected for PAO process. A cellulose-based membrane, known as a CTA membrane, was purchased from Hydration Technology Innovations (HTI, Albany, OR). Two polyamide-based membranes, generally known as TFC membranes, were purchased from HTI and Porifera Inc. (Breakwater Court Hayward, CA). Lastly, a typical polyamide-based nanofiltration (NF) membrane was also acquired from DOW-Filmtec (Minneapolis, MN). These membranes are referred to as HTI (CTA), HTI (TFC), PFO-100, and NF-90 throughout this article. All the membranes were stored at 4 °C and soaked in DI water for at least 1 h before use. It should be noted that HTI (CTA), HTI (TFC), and PFO-100 were designed for the osmotic-driven FO process, and NF-90 was designed for the pressure-driven NF system.

### 2.2. Experimental setup and operation for PAO process

A lab-scale PAO system was prepared as described elsewhere [26]. A cross-flow PAO cell was custom-built with symmetric rectangular channels (77 mm in length, 26 mm in width and 3 mm in height) on both feed and draw sides, creating an effective membrane area of 20.02 cm<sup>2</sup>. Recently researches reveal that active layer of FO membrane was deformed by the applied hydraulic pressure [24,28]. To prevent membrane breakage by the applied pressure, ten sheets of a tailored permeate carrier (Hydranautics, Inc.) were inserted in the draw

solution channel. Their effects on ICP are negligible; however, these are able to prevent unwanted deformation and/or fracture of the membranes being tested. The cross-flow velocity of co-current flows in both channels were fixed at 8.5 cm/s, using a variable speed gear pump (Cole-Palmer, Vernon Hills, IL) for draw solution recirculation and a high pressure pump (Hydracell, Minneapolis, MN) for feed solution recirculation. The temperatures of both feed and draw solutions were maintained at 25 ± 1.0 °C. The draw solution tank was placed on a digital weight scale (CAS, Korea) linked to a computer that stored the weight measurements. The feed solution conductivity was measured with a calibrated conductivity meter (Model 30, YSI Incorporated, Yellow Springs, OH).

The PAO performance tests were conducted using four different NaCl concentrations; 0.3 M, 0.6 M, 0.9 M, and 1.2 M. The PAO tests started under no additional pressure (the FO system condition) and then the hydraulic pressure was increased stepwise to 2.5 and then 5.0 bar at 30 min intervals, with the active layer facing the feed solution (AL-FS mode).

### 2.3. PAO membrane characterization

#### 2.3.1. Determination of membrane performance parameters

In order to determine the intrinsic characteristics of each membrane, three performance parameters, pure water permeability (A), solute permeability (B), and the structural parameter (S), were first measured by a two-step RO-FO method following the protocol described in previous studies [21,29]. The A and B parameters of the membranes were determined using a laboratory scale cross-flow RO test. Initially, the membranes were equilibrated with deionized water at the applied hydraulic pressure ( $\Delta P$ ) of 5.0 bar until the permeate flux reached a steady value (approximately in 15 h.). After equilibration, the volumetric permeate rate was measured at the applied pressures ranging from 1.0 to 5.0 bar with 1.0 bar increments. The water flux,  $J_w$ , at each pressure was calculated by dividing the volumetric permeate rate by the membrane area. The water permeability (A) was obtained from the slope of water flux plotted versus pressure drop.

NaCl rejection (R) was also determined at the applied pressures of 5.0 bar. Using a 10 mM NaCl feed solution, the observed rejection was determined from the difference in bulk feed ( $C_b$ ) and permeate ( $C_p$ ) salt concentrations (i.e.,  $R = 1 - C_p / C_b$ ). The solute permeability (B) was determined using Eq. (1) [12,13],

$$B = J_w \left( \frac{1 - R}{R} \right) \exp \left( - \frac{J_w}{k} \right) \quad (1)$$

in this equation,  $J_w$ ,  $k$  and  $R$  correspond to the permeate flux, mass transfer coefficient and NaCl rejection, respectively.

With the two transport parameters, A and B, obtained in the RO test, the structural parameter S is determined in the FO test. Based on Eq. (2), the modified water flux is a function of  $k$ , A, B and S; because the values of the first three are already known, S can be easily determined by measuring the initial water flux in FO experiments. Specifically, employing 0.3–1.2 M NaCl draw solution and DI water feed solution, the water flux was measured in FO mode. The membrane support structural parameter was then determined using Eq. (2)

$$J_w = A \left\{ \frac{\pi_{d,b} \exp \left( - \frac{J_w S}{D} \right) - \pi_{f,b} \exp \left( \frac{J_w}{k} \right)}{1 + \frac{B}{J_w} \left[ \exp \left( \frac{J_w}{k} \right) - \exp \left( - \frac{J_w S}{D} \right) \right]} \right\} \quad (2)$$

where  $D$  is the diffusivity of the draw solute;  $\pi_{d,b}$  is the bulk osmotic pressure of the draw solution; and  $\pi_{f,b}$  is the bulk osmotic pressure of the feed solution. Note that the diffusivity of NaCl solution was assumed to be constant between 0.3 and 1.2 M, since the variation was less than 3% [21].

To simulate FO process more closely, the A, B, and S values of each membrane were also determined under non-pressurized conditions following the one-step, so-called *single FO method* developed in previous

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