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Evaluating the viability of double-skin thin film composite membranes in forward osmosis processes

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ABSTRACT

Internal concentration polarization (ICP) and membrane fouling are two major factors impairing the performance of forward osmosis membranes. Their relative severity differs in the two operating modes of forward osmosis. The PRO mode (where the skin layer is facing the draw solution) is less susceptible to ICP, and as a result, produces a higher water flux than the FO mode (where the skin layer is facing the feed solution). Membrane fouling is severe in the PRO mode, resulting in the decline of membrane performance with time. Membrane cleaning is also more difficult in the PRO mode. There have been suggestions to add another skin layer to alleviate membrane fouling. The effects of the sandwiched membrane design on normal operations other than fouling abatement have yet to be systematically examined. In this study, the forward osmosis performance of double-skin membranes was evaluated and compared with the single-skin membranes by both theoretical calculations and experimental measurements. The results from a series of membranes with different transport properties suggested that the double-skinned membranes are not superior to the single-skinned membranes in any aspect other than their low fouling properties. Even for forward osmosis applications with serious fouling, the single-skinned membranes operating in the FO mode can still be a better alternative.

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1. Introduction

Forward osmosis is a technology with the potential to address global water shortage that deprives many regions of the world from direct access to clean and safe water supplies [1–8]. In a typical forward osmosis process, clean water is drawn from the feed solution (waste water, brackish or seawater) to the draw solution by the osmotic pressure difference across a semi-permeable membrane; pure water is then harvested by re-concentrating the diluted draw solution [1,2]. Even though the first stage of forward osmosis needs no energy input, a recent report has shown that a forward osmosis process can be more energy intensive than a reverse osmosis (RO) process because of the thermodynamic constraint in the re-concentration stage of forward osmosis [9]. Nonetheless forward osmosis is viable in applications where useful products can be produced in the first stage, such as hydration bags or fertilizer irrigation [2,10]. In addition, with the development of novel draw solutes; and the availability of low grade waste heat or renewable energy to re-concentrate the draw solution; the total energy requirement in applications such seawater desalination can be significantly reduced [11–14]. In these cases forward osmosis can still be an environmentally friendly water treatment technology.

One drawback of forward osmosis, in comparison with a pressure driven process such as RO, is the decrease in water flux due to internal concentration polarization (ICP) [15–17]. ICP is caused by the mass transfer limitations in solute transport due to the tortuosity and thickness of the membrane support, which increase the feed concentration on the support side of the skin layer in the PRO mode ("concentrative ICP"), or dilute the draw solute in the FO mode ("dilutive ICP"), as shown in Fig. 1. The effective driving force across the skin layer is therefore decreased resulting in a lower water flux. The decrease in water flux is more pronounced in FO than in PRO, as has been shown by both theoretical calculations and experimental measurements by various groups [18–21]: the dilutive ICP is always severer than the concentrative ICP.

The PRO mode, despite a higher water flux, is not the desired mode of operation if membrane fouling is significant [22–25]. Fouling is the deposition of suspended solids (or solutes) on the surface or inside the pores of a membrane, in such a way that will reduce the membrane performance. In the FO mode, foulant deposition occurs only on the surface of the skin layer which can be restored to its pristine state by cleaning. In the PRO mode, foulants

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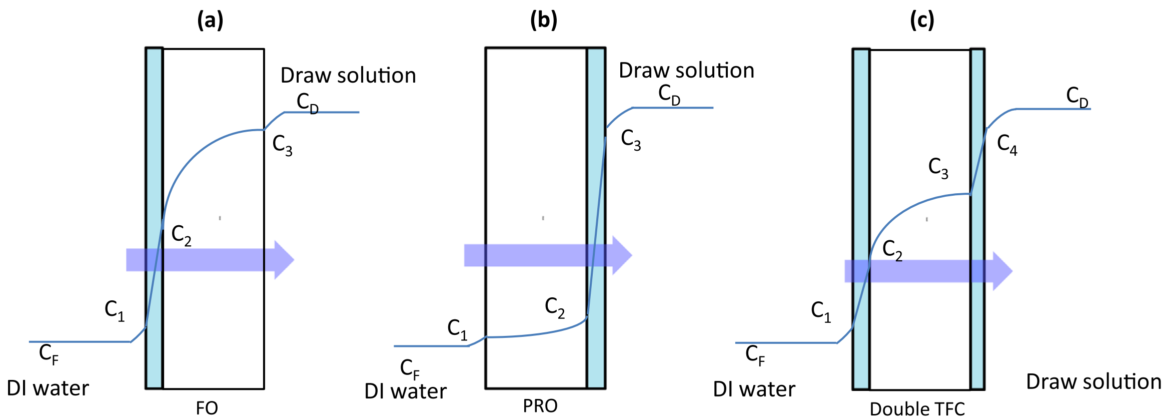

Fig. 1. The concentration profiles of the single TFC membrane in FO (a) and PRO (b) modes, and that of double TFC membrane (c).

penetrate deep into the membrane porous support layer and are more difficult to remove. On the basis of membrane maintenance against fouling, the FO mode is clearly the choice.

The membrane fouling problem in PRO can be mitigated by adding an additional skin layer on the other side of the membrane support to form a double-skin forward osmosis membrane [3,26–31]. For example, Zhang et al. prepared a double-skin cellulose acetate (CA) membrane by phase inversion, and showed easier restoration of the water flux by simple membrane cleaning after a forward osmosis operation [3]. Duong et al. deposited a layer of Nexar copolymer on the support side of a TFC membrane and reported much lower fouling propensity for emulsified oil-water separation [27]. Qi et al. deposited the double skins by a layer-by-layer technique, and demonstrated superior anti-fouling properties [28]. At first glance the double-skin design appears to provide a way to eliminate ICP since the solute now does not have direct access to the membrane support layer. However, as pointed out in the paper of Tang et al [31], where a mathematical model was used to determine the optimal combination of the skin layer transport properties at the two sides of the membrane system; ICP is still present in double-skin membranes and water flux is lower than that in single-skin membranes [31]. The model, however, did not consider the contributions from external concentration polarization (ECP) and the structural parameters of the support layer (which control the extent of ICP), and the conclusion regarding ICP is open to debate. An investigation of effects of the membrane structural parameters in double-skin vs single-skin design, with ECP included in the analysis, will address that concern. In addition, we would also like to determine any possible benefits of double-skin membranes other than an improved anti-fouling property in forward osmosis.

In this study, we present our mathematical models for both single- and double-skin membranes in forward osmosis where ECP and membrane structural factors are taken into consideration in the model development. We have also fabricated TFC membranes with polyamide skin layers formed by the interfacial polymerization (IP) of m-phenylenediamine (MPD) and trimesoyl chloride (TMC). The skin layer was deposited on either the top, or both sides of the membrane substrate (various types) to form single TFC and double TFC membranes. Forward osmosis data was collected in FO and PRO mode using the single TFC membranes, and in double thin film composite (DTFC) mode where two single TFC membranes were stacked back-to-back or a double-skin membrane was used. The models were validated first by the experimental data, and then used to predict and compare the performances between single- and double-skin membranes with different structural parameters. The predicted performance, as well as experimental measurements from a series of membranes, showed the following trend of water flux in different operating modes: DTFC < FO < PRO. Therefore for operating conditions within the boundary of this study, the DTFC mode does not offer any advantage over single TFC membrane operations other than anti-fouling protection.

2. Mathematical model development

2.1. Single TFC membranes

The mathematical models for water and salt transport through a forward osmosis membrane have been reported in a number of papers [16,31,32]. Model predictions of the experimental results are generally good after taking the substrate ICP into consideration. The ECP on the substrate side is often omitted in these models since ICP was implicitly assumed to be the dominant factor. Since this study is focused on situations of low ICP (in the DTFC mode), the ECP on both sides of the membrane may no longer be small by comparison. Hence ECP is included in the model development.

The solute concentration profile in the FO mode is shown in Fig. 1a, where the water flux (J_v, L m^{-2} h^{-1}) and reverse salt flux (J_s, g m^{-2} h^{-1}) across the membrane selective layer may be calculated from Eqs. (1) and (2) respectively, whenever the van't Hoff equation ($\pi = ncRT$) is applicable [31]:

$$J_v = A \cdot nRT(c_2 - c_1)/M \tag{1}$$

$$J_s = B \cdot (c_2 - c_1) \tag{2}$$

where A (L m^{-2} h^{-1} bar^{-1}), B (L m^{-2} h^{-1}), n, R (L bar mol^{-1} K^{-1}), T (K), c (g/L) and M (mol/g) are the water permeability coefficient, the salt permeability coefficient, the van't Hoff coefficient, the gas constant, absolute temperature, solute concentration and molecular weight of solute, respectively. The subscripts 1, 2 and 3 of the concentration terms refer to the positions of the skin layer top surface adjacent to feed, skin layer bottom surface adjacent to the support and the porous bottom layer in contact with the draw solution. The solute transport in the support layer is also governed by Eq. (3) [31]:

$$J_s = D_{eff}\frac{dc}{dx} - J_v c \tag{3}$$

with the boundary conditions:

$$c = c_2 \text{ at } x = 0 \tag{4}$$

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