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Characterization of internal and external concentration polarizations during forward osmosis processes

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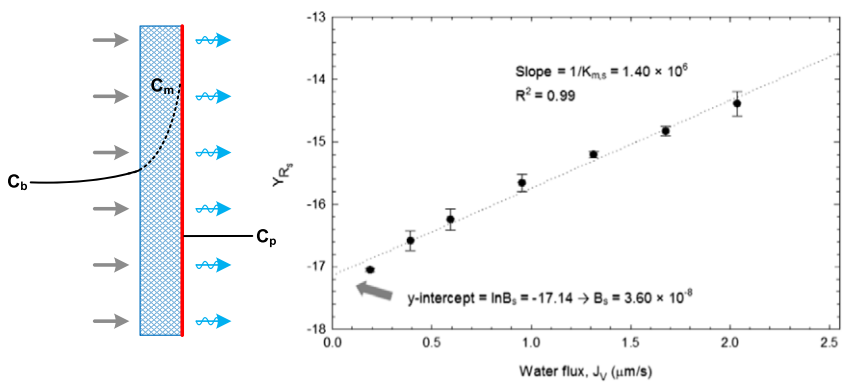
HIGHLIGHTS

- We developed direct methods for evaluating structural parameter of FO membranes using independent RO tests.
- Both internal and external CPs can be characterized simultaneously.
- The methods resulted in satisfactory prediction of FO performance.

GRAPHICAL ABSTRACT



Direct characterization of internal and external concentration polarizations

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ABSTRACT

Forward osmosis (FO) is an emerging technology showing promising future in fields of water/wastewater reclamation, desalination and energy production. However, the efficiency of such process can be adversely affected by internal concentration polarization (ICP), which is usually characterized by the structural parameter, S value. In this work, we established three independent testing methods in reverse osmosis (RO) mode to evaluate the S value, namely the J_v-method, the R_s-method and the R_t-method, relying on the measurements of RO water flux, salt rejection and trace contaminant (boron in this work) rejection, respectively. S values achieved by three methods showed good agreement with that obtained by the conventional FO water flux fitting method. Among the three approaches, the R_s-method exhibited a much smaller deviation and it was demonstrated successfully that this method can be used to estimate the value of the solute permeability graphically at the same time. In addition, the R_s-method showed the feasibility to study ICP and external concentration polarization (ECP) interpretively.

1. Introduction

Forward osmosis (FO) has been recently applied in fields of water/wastewater treatment, seawater desalination and energy harvesting, owing to its potentially low energy consumption [1,10]. However, the application of FO processes is limited by the presence of severe

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concentration polarization (CP) [1,11,15]. Previous studies revealed that the CP process occurring near FO membrane surface, known as external CP (ECP) can be attenuated by hydraulic means, such as increasing cross flow velocity [13,16,17]. However, internal CP (ICP), a unique CP phenomenon occurring inside the porous support layer of the FO membrane, is much more detrimental to the FO processes due to the difficulty to control it [18]. Severe ICP leads to the reduction of the effective driving force and thus the permeate flux [11,13,19].

Numerous studies have investigated ICP mechanisms [1,11,13,20,23], yet only a handful of studies focused on direct characterization of ICP. The structural parameter, S, has been widely used as an indicator of the extent of the ICP [13,19,24]. S is of critical importance in FO studies, and smaller S value is desirable for FO membranes due to the reduced ICP. In most of the existing FO studies, the S value is obtained indirectly by fitting the FO water flux (J_v) curves using classical ICP models (i.e., Eq. (1) for the active layer facing the feed solution (AL-FS) orientation and Eq. (2) for the active layer facing the draw solution (AL-DS) orientation [11,25])

$$J_v = \frac{D_s}{S}\left[\ln\frac{A\pi_{draw}+B_s}{A\pi_{feed}+B_s+J_v}\right](AL-FS) \tag{1}$$

$$J_v = \frac{D_s}{S}\left[\ln\frac{A\pi_{draw}+B_s-J_v}{A\pi_{feed}+B_s}\right](AL-DS). \tag{2}$$

In Eqs. (1) and (2), the membrane water permeability (A) and solute permeability (B_s) are obtained independently from filtration tests in a reverse osmosis (RO) mode. D_s is the solute diffusion coefficient. π_{feed} and π_{draw} are the osmotic pressures of the feed solution and the draw solution, respectively. This method is based on the assumption that the transport properties are universally valid for RO and FO experiments [26]. Compared to the FO J_v-fitting approach, methods that directly characterize ICP and that measure S values using independent tests are preferred. Up till now, there have been only limited attempts on the direct characterization of ICP [27,28]. In addition, the existing FO J_v-fitting method does not allow independent assessment of ICP vs. ECP effects. Despite that ECP may play a significant role in FO [29,33], to the best knowledge of the authors, direct characterization of ECP in FO process is still lacking.

The objective of this study was to develop direct approaches to characterize FO membrane support structures. In this work, independent RO testing procedures were established to evaluate the structural parameter of FO membranes. Three approaches, relying on RO water flux measurements (the J_v-method), salt rejection measurements (the R_s-method) and trace contaminant (boron in the current study) rejection measurements (the R_t-method), were developed and compared. Particularly, the R_s-method provided a convenient solution to determine both the structural parameter of the support layer and the separation properties of the rejection layer. Moreover, this method was feasible to resolve the effect of ECP from that of ICP experimentally.

2. Theory

2.1. RO water flux method (J_v-method)

In a conventional RO test, a membrane is placed with its active layer facing the concentrated feed solution and the support layer (SL) facing the permeate side (Fig. 1(a)). Such membrane orientation is denoted as SL-permeate in the current study. The membrane water flux J_v is described by the classical solution-diffusion model [11,34]

$$J_v = A(\Delta P - \Delta\pi) \tag{3}$$

where ΔP represents the hydraulic pressure difference across the membrane active layer and $\Delta\pi$ is the osmotic pressure difference between the membrane surface π_m and the permeate stream π_p.

In the SL-permeate orientation, ICP is not important as there is no solute accumulation in the membrane porous support. However, ICP does occur when the membrane orientation is reversed in an RO test, i.e., the support layer is facing the feed solution (SL-feed, see Fig. 1(a)). In this orientation, the solutes in the feed water entering the porous support layer are rejected by the membrane active layer, leading to a concentration build-up inside the support. Consequently, the osmotic pressure inside the support is increased and the permeate flux is decreased. In addition to ICP, ECP occurring next to the porous layer where ICP occurs, may also have a negative effect on the permeate flux. By considering both ECP and ICP and applying the boundary layer film theory [35], we have:

$$\frac{C_m - C_p}{C_b - C_p} = e^{J_v/k_{eff}} \tag{4a}$$

where C_b, C_m, C_p are the concentrations of the bulk feed solution, the membrane surface and the permeate water, respectively. k_{eff} is the effective mass transfer coefficient, given by the ratio of effective solute diffusion coefficient D_{eff}, to the effective boundary layer thickness δ_{eff} ($k_{eff} = D_{eff}/\delta_{eff}$). Here the effects of both ICP ($K_{m,s}$, mass transfer coefficient of the solute in the porous support layer) and ECP (k_s, solute mass transfer coefficient outside the support) are lumped into a single term k_{eff}, holding the relationship that $1/k_{eff} = 1/K_{m,s} + 1/k_s$. By further assuming that the osmotic pressure is proportional to the solute concentration [13,36],

$$\frac{\pi_m - \pi_p}{\pi_b - \pi_p} = e^{J_v/k_{eff}} \tag{4b}$$

where π_b, π_m, π_p are the osmotic pressures of the bulk feed solution, the membrane surface and the permeate water, respectively. From Eqs. (4b), (3) can then be expressed as:

$$J_V = A\left[\Delta P - e^{J_v/k_{eff}} \cdot \left(\pi_b - \pi_p\right)\right]. \tag{5}$$

It is worthwhile to note that Eq. (5) is derived based on the assumption of linear concentration–osmotic pressure relationship, which may not be valid for some concentrated solutions [37] (see further discussion in Sections 4.1.1 and 4.1.4).

By re-arranging Eq. (5), we can have the following relationship:

$$\ln\left(\frac{\Delta P - \frac{J_v}{A}}{\pi_b - \pi_p}\right) = \frac{J_v}{k_{eff}}. \tag{6}$$

By plotting experimental $\ln[(\Delta P - J_v/A)/(\pi_b - \pi_p)]$ values as a function of J_v, k_{eff} can be readily determined as the reciprocal of the slope of the curve.

Eq. (6) is also applicable to the SL-permeate orientation by setting $1/k_{eff} = 1/k_s$ (i.e., no ICP effect). Thus, one can determine the external mass transfer coefficient k_s in the SL-permeate orientation (ECP effect only) and the overall mass transfer coefficient in the SL-feed orientation (combined ICP and ECP effects). By comparing the two membrane orientations, the ICP related mass transfer coefficient $K_{m,s}$ can be resolved. Alternatively, one can obtain $K_{m,s}$ directly from the SL-feed tests by applying a relatively high cross flow velocity to minimize the effect of ECP (see Section 4.2). By further assuming that the osmotic pressure of the permeate water is much less comparing to that of the feed solution (i.e., high membrane rejection), we can have:

$$\ln\left(\Delta P - \frac{J_v}{A}\right) = \frac{J_v}{K_{m,s}} + \ln\left(\pi_b - \pi_p\right) \approx \frac{J_v}{D_s/S} + \ln\pi_b \tag{7}$$

where D_s is the solute diffusion coefficient inside the porous support layer. S ($=\tau_m \cdot l_m/\varepsilon_m$) is the structural parameter of the support layer