



Rejection of nine haloacetic acids and coupled reverse draw solute permeation in forward osmosis



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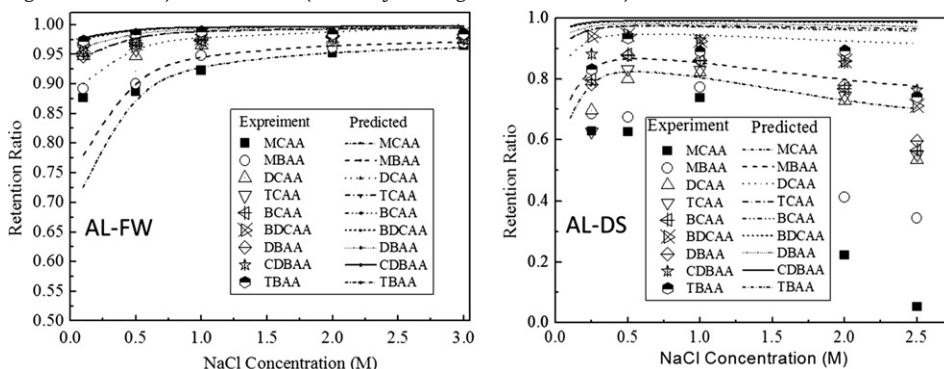
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HIGHLIGHTS

- Forward osmosis rejects haloacetic acids (HAAs) well.
- The active-layer facing feed water (AL-FW) orientation is preferred.
- The solution-diffusion model predicts HAA rejection better for AL-FW.
- The solution-diffusion model accurately predicts reverse draw solution permeation.

GRAPHICAL ABSTRACT

The behaviors of haloacetic acid (HAA) rejection by forward osmosis are obtained by experiments and predicted by the solution-diffusion model combined with concentration polarization for both the AL-FW (active layer facing the feed water) and the AL-DS (active layer facing the draw solution) orientations.



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ABSTRACT

The rejection of haloacetic acids (HAAs) by forward osmosis (FO) and the coupled reverse draw solute permeation were experimentally determined and mathematically modeled by using the solution-diffusion model for both the AL-FW (active layer facing the feed water) and the AL-DS (active layer facing the draw solution) orientations. The rejection ratio for each HAA increased with the increase of draw solute concentration for the AL-FW orientation. In contrast, the HAA rejection ratio could reach its maximum under a medium osmotic pressure difference for the AL-DS orientation. The rejection ratios for all HAAs were higher than 94.6% for the AL-FW orientation and ranged from 73.8% to 89.1% for the AL-DS orientation under a draw solute concentration of 1 mol/L NaCl. The reverse draw solute flux for the AL-FW orientation was lower than that for the AL-DS orientation. The model-predicted HAA rejection results matched well with the experimental rejection ratios for the AL-FW orientation. However, the model over-estimated the rejection ratios for the AL-DS mode, likely due to the adoption of inaccurate mass transfer coefficient for internal concentration polarization. Regarding the reverse draw solute permeation, a general agreement between the model prediction and experimental data was observed for both orientations.

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1. Introduction

Forward osmosis (FO) is a process driven by the difference in chemical potentials across a semi-permeable membrane with little hydraulic pressure applied. In recent years, the potential application of FO to

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wastewater reclamation has been extensively studied [1–3], which was normally fulfilled by combining FO with other technologies as a hybrid process [4]. The hybrid processes may compete favorably with the simple pressure-driven processes such as reverse osmosis (RO) owing to the fact that the former may have a lower demand on electrical energy and its membranes may suffer less membrane fouling [2,5–7]. One of the biggest concerns about wastewater reclamation is the removal efficiencies for the large number of trace organic compounds at concentrations in the magnitudes of ng/L to $\mu\text{g/L}$. Common trace organic compounds include endocrine disruptors (EDCs), pharmaceutical and personal care products (PPCPs) and disinfection by-products (DBPs) [8–11]. The reclaimed water is better to be free of these trace organic compounds due to their potentially high health risks to humans and aquatic organisms.

Previous studies [2,12–17] showed that the rejection of PPCPs and EDCs by FO process varied among the compounds and the operational conditions. Results by Hancock revealed that rejection efficiencies by FO depended primarily on molecular size and charge. Rejection of positively and negatively charged trace organic compounds was greater than 80%, while rejection of the nonionic trace organic compounds varied, between approximately 40% and 90% [18]. Xie et al. [13] found that the membrane orientation (AL-FW (active layer facing feed water) and AL-DS (active layer facing draw solution)) affected the rejection performance.

Development of mathematical models capable of predicting the rejection of the trace organic compounds by the FO process will improve the economic efficiency of the process and expand its applications. By far, there has been only a few work conducted on the modeling of the rejection performance by FO, among which the model based on the solution-diffusion model developed by Jin et al. [19] was found able to accurately predict the FO rejection of inorganic solutes. It is also desirable to model the permeate water flux and the reverse draw solute flux through the membrane. The limited water flux and the substantial reverse draw solute flux are among the biggest impediments to the viability of the FO processes [20,21]. In this regard, a few studies investigated the reverse draw solute permeation in the FO process and proposed a relationship with the water flux [22,23]. Better understanding and prediction of feed contaminant and coupled reverse draw solute permeation in FO membrane processes can facilitate to optimize the operational conditions and benefit to fabricate FO membranes that minimize the loss of draw solute into the feed solution, thereby reducing operating costs.

Haloacetic acids (HAAs) are among the DBPs with the highest concentrations in chlorinated or chloraminated sewage treatment plant effluent. The trichloroacetic acid (TCAA) concentration can be as high as $471 \mu\text{g/L}$ in the chlorinated wastewater effluent [24]. This study mainly investigated the rejection of nine HAAs and the reverse draw solute permeation in the FO process for both the AL-FW and the AL-DS orientations. Substantial attention was paid to the mathematical description of the water, HAA and draw solute fluxes. This study also evaluated the feasibility of the solution-diffusion model in predicting the rejection of organic contaminants and coupled draw solute permeation in the FO process.

2. Materials and methods

2.1. The FO system and operations

The schematical diagram of the bench-scale FO system is shown in Fig. 1. The membrane unit was custom built with two flow channels of 2 mm in height both, which were separated by an FO membrane coupon that had an effective area of 40.5 cm^2 . The commercial FO membrane was provided by Hydration Technologies, Inc. (Albany, OR) and, according to the manufacturer, was made of cellulose triacetate supported by embedded polyester screen mesh. The recirculation flows of the feed water (FW) and the draw solution (DS) on the two sides of the

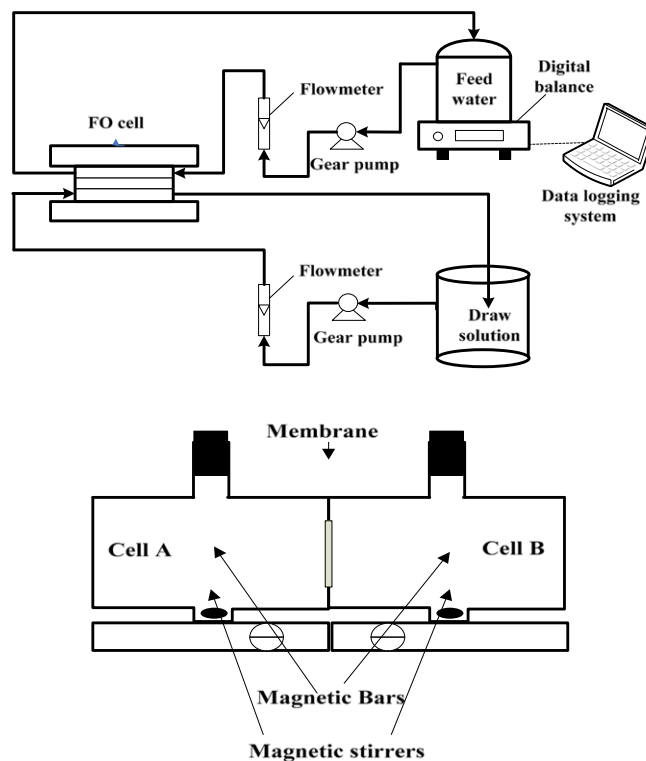


Fig. 1. Schematic diagrams of the bench-scale forward osmosis system (top) and the diaphragm cell test unit (bottom).

membrane were counter-current with a flow velocity of 20.4 cm/s both, each regulated by a variable-speed gear pump (Longer, USA). The relatively high cross-flow velocity can substantially suppress the external concentration polarization (ECP) that will be described in detail later.

Ultrapure water (Milli-Q, Millipore, USA) was used for the FW and the DS preparation. The FW was prepared by dissolving a mixture of the nine haloacetic acids (HAAs) (Sigma-Aldrich, Germany) into a 10 mmol/L NaCl (as background solute) solution with the final concentration for each HAA at $200 \mu\text{g/L}$. The FW ionic strength was similar to the typical values for freshwater including sewage treatment plant effluent, although our preliminary study showed that the FW ionic strength ($0\text{--}20 \text{ mmol/L}$) had negligible influence on the rejection of HAAs. The nine HAAs were monochloroacetic acid (MCAA), monobromoacetic acid (MBAA), dichloroacetic acid (DCAA), bromochloroacetic acid (BCAA), dibromoacetic acid (DBAA), trichloroacetic acid (TCAA), bromodichloroacetic acid (BDCAA), dibromochloroacetic acid (CDBAA) and tribromoacetic acid (TBAA). Physicochemical properties for each HAA including molecular weight (MW) and dissociation constant (pK_a) were summarized in Table 1. Sodium bicarbonate was added into the FW to adjust the pH to neutral ($\text{pH} = 7$). No natural organic matter (NOM) or soluble microbial products (SMPs) were contained in the FW, though both NOM and SMP could not only adsorb certain trace organic compounds but also lead to membrane fouling, which would greatly affect the rejection performance of the FO process [12]. The DS was a NaCl solution at a concentration varying from 0.1 to 3 mol/L .

The FO experiments were conducted in an air-conditioned room with the temperature set at $25 \pm 1 \text{ }^\circ\text{C}$. Each FO test lasted for about 4 h. The duration was long enough to obtain accurate water, HAA and reverse draw solute permeation fluxes. On the conclusion of each FO test, the volume variation of the DS (or the FW) was less than 5%, and as such the draw solute (in the DS) and the HAA (in the FW) concentration variation were negligible. The FW tank was placed on a digital scale (Mettler Toledo, Germany) connected to a computer data logging

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