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Effect of reverse permeation of draw solute on the rejection of ionic nitrogen inorganics in forward osmosis: Comparison, prediction and implications

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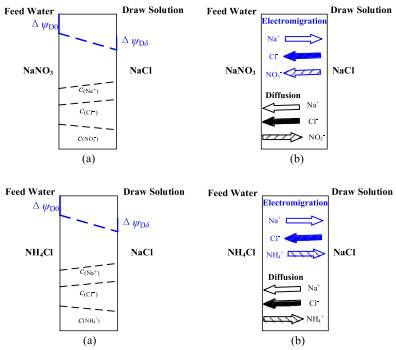
GRAPHICAL ABSTRACT



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ABSTRACT

In this study, the influence of reverse draw solute permeation on the rejection and prediction of NO_3^- and NH_4^+ by forward osmosis (FO) were investigated. The rejection of NO_3^- and NH_4^+ were experimentally compared between FO mode (active layer facing the feed water) and RO mode (hydraulic pressure applied). Under similar water fluxes, the rejection ratio of NO_3^- by HTI-ES membrane was much higher during RO than it was during FO, while the opposite was true for HTI-TFC membrane. For both membranes, the rejection of NH_4^+ was higher during RO than that it was in FO. Rejection was mathematically predicted by using the solution–diffusion model with the permeability obtained from the diffusion cell. The predicted NH_4^+ rejection for the HTI-TFC membrane matched well with the experimental rejection ratios. However, the model greatly overestimated the rejection ratios of NO_3^- by HTI-ES membrane and underestimated the rejection of NH_4^+ by HTI-ES membrane and NO_3^- by HTI-TFC membrane. The electrostatic equilibrium at the interface and electrostatic gradient across the membrane may differ between RO, FO and diffusion cell mode, which should be considered in future FO models.

1. Introduction

Forward osmosis (FO) is a promising water reclamation and sludge dewatering technology as it can simultaneously purify water and recover resources (carbon, ammonia and phosphorus in the wastewater) [1–4]. However, the removal efficiency or recovery capability of common nutrients (i.e., NH_4^+ or its nitrification product NO_3^-) in wastewater and sludge has always received great attention from researchers worldwide [5,6].

The rejection mechanisms and behaviors of FO share some characteristics with pressure driven processes (i.e., NF and RO) and both can be predicted by the solution-diffusion model. The rejection efficiency of

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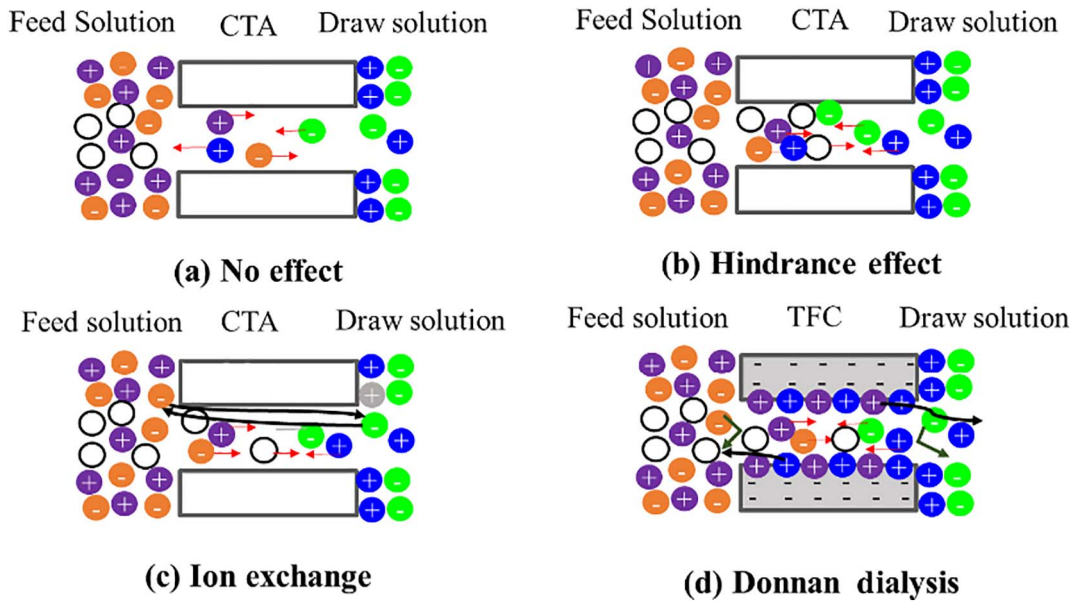

Fig. 1. The influence and mechanisms of draw ions permeation on the transport of feed contaminants.

FO is similar to that of RO, however, the unique reverse permeation of the draw solute during FO could affect contaminant rejection in the feed water (FW). Four scenarios can be observed in previous studies by comparing the rejection ratios with RO mode or the prediction results (with the permeability coefficient obtained from the RO mode) [7–11]. Jin et al. [7] and Kong et al. [11] found that the solution-diffusion model could accurately predict the rejection of inorganic solutes and haloacetic acid by cellulose triacetate (CTA) FO membrane, demonstrating that the reverse permeation of FO had no influence on the rejection of these compounds (Fig. 1a). Meanwhile, Xie et al. [8] found that the rejection of some hydrophobic trace organics was higher during FO than it was during FO than it was during RO because forward diffusion was hindered by the reverse permeation of the draw solute (Fig. 1b). However, Hancock et al. [9] found that strong electrolyte ion fluxes excluding nitrate, through the CTA membrane can be well predicted by the solution-diffusion model, which is attributed to an ion exchange mechanism that allows the nitrate in the feed solution to exchange rapidly with anions from the draw solution, resulting in a lower nitrate rejection for FO than RO mode (Fig. 1c). Kong et al. [10] also observed a similar phenomenon that the rejection ratios of some negatively charged pharmaceuticals were much lower in FO mode than RO mode for a CTA membrane. However, Lu et al. [12] and Arena et al. [13] proposed that Donnan dialysis enhanced the bidirectional diffusion of cations through polyamide thin film composite (TFC) membranes, which could greatly enhanced the transport of positively charged ions and result in lower rejection ratios (Fig. 1d). In conclusion, the rejection observations and mechanisms of rejection depend on contaminants, membranes and operational modes. The transport mechanisms and model of FO process may not be the same as those in RO due to the reverse permeation of draw solute [14,15].

The additional mechanism (i.e., hindrance effect, ion exchange and Donnan dialysis) induced by the reverse permeation of draw solute could affect the forward transport of solutes in the feed, potentially resulting in inadequate solute retention and a decrease in the water-solute selectivity (Fig. 1) [16,17]. It should also be noticed that diffusion cell tests are often adopted to determine the permeability coefficient (B) - of solute in FO process, but the absence of reverse permeation in diffusion cell could influence the prediction accuracy [10,18]. Therefore, a re-examination and a comprehensive study of ion rejections and its associated mechanisms, and prediction accuracy by using solution-diffusion model with the permeability coefficient obtained from the diffusion cell test are required to gain a fundamental understanding on the mass transfer in FO.

The objectives of this work were: (1) to compare the NH_4^+ and NO_3^- rejection ratios for two membranes during FO and RO, (2) to systematically investigate and analyze the bi-directional diffusion of ions across FO membrane, (3) to examine the role of reverse draw solute permeation on the prediction of NH_4^+ and NO_3^- with the permeability coefficient obtained from diffusion test and (4) to explore transport mechanism during the FO process and the potential implications for developing a FO model. Understanding these will benefit the development of FO membranes with high rejection of NH_4^+ and NO_3^- and improve FO models.

2. Materials and methods

2.1. Membranes

Two commercial FO membranes (HTI-ES and HTI-TFC) used in this study were both obtained from Hydration Technologies, Inc. (Albany, OR, US), and were respectively made of CTA and polyamide. The membrane has been comprehensively characterized in previous studies [10,19]. Prior to the RO experiments, the membranes were soaked in ultrapure water (Millipore, US) for at least 12 h. Prior to FO experiments, the HTI-TFC membrane was first soaked in a 50% solution of isopropyl alcohol for 5 min and then stored in ultrapure water to maintain hydration [20], while the HTI-ES membrane was soaked for at least 12 h in ultrapure water at room temperature.

2.2. The RO system and operations

A cross-flow RO system, which has been described in our previous studies [10,11], was used to determine the water permeability coefficient (A), NaCl permeability coefficient (B_{NaCl}) and rejection ratios of NH_4^+ and NO_3^- under different hydraulic pressure with the cross-flow velocity of 30.4 cm/s and the temperature of 25 ± 1 °C.

The virgin membranes were first compacted by filtering by filtering high-purity water (Milli-Q, Millipore, USA) at 13 bar until a stable water flux was reached. The pure water and 10 mmol/L NaCl were employed as the feed water to obtain the A and B values for the two membranes with the pressure appropriately from 3 to 11 bar, respectively. The A and B value can be calculated by $A = J_w/(\Delta P - \Delta\pi)$ and