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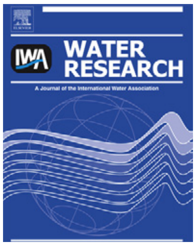

Combined organic–inorganic fouling of forward osmosis hollow fiber membranes

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ABSTRACT

This research focused on combined organic–inorganic fouling and cleaning studies of forward osmosis (FO) membranes. Various organic/inorganic model foulants such as sodium alginate, bovine serum albumin (BSA) and silica nanoparticles were applied to polyamide-polyethersulfone FO hollow fiber membranes fabricated in our laboratory. In order to understand all possible interactions, experiments were performed with a single foulant as well as combinations of foulants. Experimental results suggested that the degree of FO membrane fouling could be promoted by synergistic effect of organic foulants, the presence of divalent cations, low cross-flow velocity and high permeation drag force. The water flux of fouled FO hollow fibers could be fully restored by simple physical cleaning. It was also found that hydrodynamic regime played an important role in combined organic–inorganic fouling of FO membranes.

1. Introduction

Forward osmosis (FO) membrane process is a relatively new separation technology. Unlike pressure-driven membrane processes, the driving force for FO arises from the osmotic pressure gradient between the draw and the feed solutions separated by a membrane. FO process has several potential advantages of lower energy demand and less prone to membrane fouling as no external pressure is applied.

Nowadays FO process attracts an increasing interest as emerging technique for desalination. Despite the fact that the composition of saline water varies widely from source to source, all substances present in saline water can be divided into two groups: organic and inorganic. Hence, studies on combined organic–inorganic fouling of FO membranes are of great significance. Most research works on FO, however, are dedicated to the investigation of fouling caused by either one-component (Lay et al., 2010; Lee et al., 2010; Mi and Elimelech, 2008, 2010; Tang et al., 2010; Wang et al., 2011; Wang et al., 2010b; Zou et al., 2011) or complex feed solution (wastewater, seawater, etc.) (Achilli et al., 2009; Cornelissen et al., 2008; Garcia-Castello and McCutcheon, 2011; Holloway et al.,

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2007; Lay et al., 2012; Lutchmiah et al., 2011; Qin et al., 2010; Yangali-Quintanilla et al., 2011; Zhang et al., 2012; Zhao and Zou, 2011; Zhao et al., 2011). Former studies on single component fouling in FO membranes have reported that the fouling behavior was strongly affected by foulant properties (type, size and concentration), feed solution pH, divalent cations, membrane orientation, cross-flow velocity, spacer, draw solution type and concentration.

Despite its importance, there are only limited works that discuss combined fouling, both in reverse osmosis (RO) and FO processes. Combined organic–organic (Ang and Elimelech, 2007; Ang et al., 2011; Li et al., 2007; Wang and Tang, 2011) and organic–inorganic (Higgin et al., 2010; Zhu and Elimelech, 1995) fouling had been investigated in RO systems. The findings indicated that membrane performance could be greatly affected by a synergistic effect between foulants. BSA-lisozyme, BSA-alginate and humic acid-aluminum oxide aggregates could synergistically enhance fouling of RO membranes (Ang et al., 2011; Li et al., 2007; Wang and Tang, 2011; Zhu and Elimelech, 1995). On the contrary, Higgin et al., 2010 reported that specific interactions between alginic acid and silica could mitigate fouling. Nayak et al., 2011 compared FO water fluxes of various model systems (feed containing a combination of sucrose, dextrose and pectin) in different membrane orientations. Using natural seawater as feed, Li et al., 2011 suggested that fouling of FO flat sheet membrane could be promoted by reaction between dissolved silica and natural organic matter. Further to this finding, a more recent work reported that the FO flux decline of feed containing a mixture of small and large silica nanoparticles was more prominent than the flux drop of a monodisperse colloidal suspension (Boo et al., 2012). A more cohesive cake layer was formed by polydisperse silica nanoparticles as compared to the cake formed by only large particles. Hence, a low porosity cake layer would significantly limit the back diffusion of salts that increased cake-enhanced osmotic pressure.

Our present work aims to investigate a combined organic–inorganic fouling of FO membranes. Bovine serum albumin (BSA), alginate and silica nanoparticles were used as model foulants. Experiments were performed by using FO hollow fiber membranes fabricated in our laboratory with the active layer of the membrane facing the feed solution (AL-FS). Interactions between single, binary and ternary components with the membrane were examined. The effects of osmotic driving force, divalent cations, cross-flow velocity and hydrodynamic operating regime on membrane fouling and fouling reversibility were also investigated.

2. Materials and methods

2.1. FO membranes and setup

In-house fabricated FO hollow fiber membranes were employed in this study. The characteristic parameters of the membrane are summarized in Table 1. The hollow fiber FO setup used for filtration experiments was similar to that reported by Wang and coworkers (Chou et al., 2010; Wang et al., 2010a) with the membrane area of 122 cm^2.

Table 1 – Membrane characteristics.

Parameter	
Configuration	Hollow fiber
Active layer material	Polyamide
Support layer material	Polyethersulfone
Thickness, μm	183
Support mean pore size, nm	12.7[a]
Contact angle of support layer,	78[a]
Water permeability (A), L/m^2-h-atm	2.8
NaCl permeability (B), L/m^2-h	0.17
NaCl Rerejection (R), %	93
Mass transfer coefficient, m/s	2.5×10^{-6} [a]
Structural parameter, m	5.95×10^{-4} [a]

a Wang et al., 2010a.

2.2. Measurements of intrinsic separation properties of the membrane

Prior to fouling experiments, the intrinsic separation properties, i.e. pure water (A) and salt (B) permeability, as well as the salt rejection (R) of the membranes were measured. The tests were performed under cross-flow condition with a flow rate of 450 ml/min at 1 bar and room temperature (23 ± 1 °C). The salt rejection was measured by using 500 ppm NaCl as feed water (Fang et al., 2012). The salt permeability was calculated based on solution-diffusion theory as follows:

$$R = \left(1 - \frac{B}{A(\Delta P - \Delta\pi)}\right)^{-1} \tag{1}$$

where ΔP and $\Delta\pi$ are the applied and osmotic pressure differences across the membrane, respectively.

2.3. FO experiments

Continuous FO experiments were carried out in the AL-FS orientation for 6 h. Baseline tests were performed to evaluate the FO flux behavior without the presence of foulants. The feed solution for baseline tests contained 10 mM NaCl with pH of 6.25. The feed solution used for fouling experiments was a mixture of alginate (Hayashi Pure Chemical Industries Ltd., Japan), BSA (Sigma–Aldrich, USA) and silica colloids of 10–20 nm (Sigma–Aldrich, USA) in the presence or absence of Ca^{2+} ions. The working volume of the feed solution in the tank was 4.5 L. Various concentrations of 0.5, 1, 3 and 4 M NaCl were utilized as draw solutions. The concentration of draw solution was maintained constant throughout the experiment by using 5 M NaCl dosing solution. The FO experiments were conducted at 6, 12 or 24 cm/s cross-flow velocity (the same for both feed and draw solutions) in countercurrent flow configuration (the feed and draw solutions were flowing tangential to the membrane surface, but in opposite directions). The permeate flux was determined at fixed time intervals by measuring the weight changes of the feed solution with a digital mass balance (Mettler Toledo, Switzerland) connected to a computer data logging system. The experiments were replicated to ensure the reproducibility of results. The water flux profiles were plotted by taking the average values obtained from several FO filtration