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Organic fouling of forward osmosis membranes: Fouling reversibility and cleaning without chemical reagents

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ABSTRACT

The recently resurgent forward osmosis (FO) membrane process has the potential to become a sustainable alternative to conventional membrane processes. However, the fouling and cleaning behavior of FO membranes remains largely unknown. There is a need to fully understand the fouling phenomena in FO in order to take advantage of this emerging technology. In this study, we used alginate as a model organic foulant to examine FO membrane fouling and cleaning behavior with the ultimate goal of determining the underlying FO fouling/cleaning mechanisms. Results showed that alginate fouling in FO is almost fully reversible, with more than 98% recovery of permeate water flux possible after a simple water rinse without any chemical cleaning reagents. We also studied the role of applied hydraulic pressure in membrane fouling and cleaning by performing fouling tests in FO (without hydraulic pressure) and RO (with hydraulic pressure) modes. Flux recovery in the FO mode was much higher than that in the RO mode under similar cleaning conditions, although the rate of membrane flux decline was similar in the two modes. The fouling reversibility of FO was attributed to the less compact organic fouling layer formed in FO mode due to the lack of hydraulic pressure. Our results suggest that operating in FO mode may offer an unprecedented advantage in reducing or even eliminating the need for chemical cleaning. AFM force measurements were used to elucidate the impact of membrane materials (cellulose acetate versus polyamide) on membrane fouling and cleaning behavior. Adhesion force data revealed that a small percentage of relatively adhesive sites on the membrane surface play an important role in increasing membrane fouling potential and decreasing cleaning efficiency. This finding implies that using average adhesion force to predict membrane fouling potential is inadequate. Extensive long-range adhesion forces are observed for the polyamide membrane in the presence of alginate and calcium ions. The long-range interactions are attributed to calcium bridging of alginate molecules between the AFM probe and the adhesive sites on the polyamide membrane surface.

1. Introduction

Water scarcity is one of the most challenging problems affecting people throughout the world [1,2]. Membrane technology, since its initial development in the 1960s, has evolved rapidly and will continue to expand as an effective approach to addressing water shortages. To date, the most widely used membrane processes in water purification applications include microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO). These membrane processes use hydraulic pressure as the driving force for water flux. However, the productivity of these pressure-driven membrane processes is severely hampered by the long-standing problem of fouling.

Numerous studies have been conducted to elucidate the mechanisms governing the fouling of pressure-driven membranes and to alleviate fouling by, for example, pretreating feed water, improving the antifouling properties of membranes, and optimizing operating conditions [3–7]. Despite these efforts, fouling still remains a major obstacle for efficient use of virtually all pressure-driven membrane systems. Thus, frequent maintenance activities, such as the commonly employed chemical cleaning, are necessary to maintain consistent membrane performance. Chemical cleaning, however, not only impairs membrane selectivity and shortens membrane life but also, is unsustainable as it consumes additional energy and produces concentrated waste streams [8,9]. Therefore, alternative membrane processes that can reduce or even eliminate chemical cleaning are strongly desired.

A recently resurgent membrane process, forward osmosis (FO), is a potential, sustainable alternative to conventional pressure-driven membrane processes [10]. FO as a concept dates back to the 1970s [11], and does not require hydraulic pressure for its

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operation. Instead, this process uses a concentrated draw solution to generate high osmotic pressure, which pulls water across a semi-permeable membrane from the feed solution. The draw solute is then separated from the diluted draw solution in order to be recycled, thereby producing clean product water. This osmotically driven process has a number of salient advantages. For example, FO could potentially consume only about 20% of the electrical energy required by other desalination processes and it may be able to use very low quality heat, such as waste heat from power plants, as its energy input [12]. In addition, FO may also have higher recovery, resulting in less brine discharge to the environment [13]. Promising FO applications include seawater desalination [11], green power production [14–16], wastewater reclamation [13,17,18], industrial wastewater treatment [19], and liquid food processing [20,21].

Despite these benefits of the FO process, its applications in real-world water purification have long been hindered by three challenging problems. First, draw solutions that are able to create high osmotic pressure using solutes that can be removed efficiently and completely have not been easily identified. Second, membranes designed specifically for osmotically driven FO processes are needed, as commercially available semi-permeable membranes are primarily designed for pressure-driven processes. Third, compared to abundant research that has been conducted on pressure-driven membrane processes, there lacks a systematic mechanistic understanding of the fouling and cleaning behaviors of FO processes. As a result, advances in FO technologies lag far behind those in pressure-driven membrane technologies.

Until recently, very few studies on FO fouling have been reported in the literature [19,22–24]. As a result, the underlying fouling/cleaning mechanisms of FO processes are basically unknown. There are several fundamental questions that remain to be answered, including (i) whether the lack of hydraulic pressure in FO processes mechanistically alters the membrane's fouling and cleaning behavior and (ii) whether membrane materials affect the fouling and cleaning behavior and which factors should be considered in the development of a new FO membrane. Therefore, fundamental and systematic research is immediately needed to elucidate the fouling/cleaning mechanisms of FO membranes.

This study examines the organic fouling and subsequent cleaning behavior of FO membranes with the ultimate goal of better understanding the underlying FO fouling/cleaning mechanisms. Alginate is used as a model organic foulant to investigate FO membrane fouling reversibility. We also study the role of hydraulic pressure in membrane fouling and cleaning by comparing membrane performances in FO (without hydraulic pressure) and RO (with hydraulic pressure) modes. Finally, we employ AFM force measurements to identify the role of membrane materials in determining membrane fouling and cleaning behavior.

2. Materials and methods

2.1. Organic foulant (alginate)

Sodium alginate was used as a model organic foulant (Sigma–Aldrich, St. Louis, MO). Alginate has been extensively used in membrane fouling research to represent polysaccharides that constitute a major fraction of soluble microbial products in wastewater effluent [25,26]. According to the manufacturer, the sodium alginate has a molecular weight in the range of 12–80 kDa. Other properties of the alginate used are given in our earlier publications [27]. A stock solution of alginate (10 g/L) was prepared by dissolving sodium alginate (received in powder form) in deionized (DI) water and mixing for over 24 h to ensure complete dissolution. The stock solution was stored in sterilized glass bottles at 4 °C without further purification.

2.2. FO membranes

The FO membrane was obtained from Hydration Technologies, Inc. (Albany, OR). The membrane has an asymmetric structure and is made of cellulose acetate (CA) supported by embedded polyester mesh. The total thickness of the membrane is approximately 50 μm. Other characteristics of the membrane are given in McCutcheon et al. [28]. A thin-film composite polyamide (PA) membrane customized for FO purposes (Dow Chemical Company, Midland, MI) was used to study the effects of membrane materials on membrane fouling and cleaning behavior. The proprietary membrane is likely modified from a commercial seawater RO membrane (Filmtec SW30XLE-400i) by reducing the thickness of the membrane support layer. The CA and PA membranes have similar pure water permeabilities determined in RO mode: 3.6×10^{-12} and 3.8×10^{-12} m/(s Pa) for the CA and PA membranes, respectively.

2.3. Membrane test systems

The fouling and cleaning experiments were conducted in two separate bench-scale membrane crossflow systems: one FO system and one RO system. The FO system comprises a custom-built crossflow membrane cell with equally structured rectangular channels on both sides of the membrane. The dimensions of the channels are 77 mm long, 26 mm wide, and 3 mm deep. No spacers were used in the channel to accelerate membrane fouling. Two variable speed gear pumps (Micropump, Vancouver, WA) were used to generate crossflows, forming separate closed loops for the feed and draw solutions. The draw solution tank was placed on a digital scale (Denver Instruments, Denver, CO) and weight changes were monitored by a computer to record the permeate flux. A constant feed and draw solution temperature of 20 ± 1 °C was maintained by a water bath (Neslab, Newington, NH). Details on the FO membrane unit are given in our recent publication [24].

The RO system consists of a custom-built plate-and-frame membrane cell with a rectangular feed channel having dimensions of 77 mm in length, 26 mm in width, and 3 mm in depth. A Hydra-cell pump (Wanner Engineering, Inc., Minneapolis, MN) was used to generate crossflow, and a bypass valve and backpressure regulator were used to control the crossflow velocity and hydraulic pressure. The permeate flux was monitored continuously by a digital flow meter (Optiflow 1000, Humonics, CA) interfaced with a computer. The temperature was maintained at 20 ± 1 °C with a water bath (Neslab, Newington, NH).

2.4. Membrane fouling and cleaning protocols

Fig. 1 illustrates the protocols for the fouling and cleaning experiments. Before each fouling experiment, a new membrane coupon was installed in the membrane unit and stabilized to obtain a constant flux. All the fouling and cleaning experiments performed in this study are operated in FO mode, where the membrane dense (active) layer faces the feed solution. The stabilization process, not shown in Fig. 1, takes about 1 h for FO and 5 h for RO. The membrane in the FO mode was stabilized with DI as the feed and 4 M NaCl as the draw solution. The membrane in the RO mode was stabilized/compacted under 31 bars (450 psi) hydraulic pressure with DI as the feed.

As shown in Fig. 1, the pure water flux of the stabilized membrane was obtained before the fouling experiment. The flux in the FO mode was obtained with DI water as the feed solution and 4 M NaCl as the draw solution. The flux in the RO mode was obtained with DI water as the feed solution and 28 bars (400 psi) applied hydraulic pressure. Following this step, the fouling experiment was performed for 20–24 h. Experimental conditions for the fouling experiments in the FO mode include: draw solution containing