



# Combined organic and colloidal fouling in forward osmosis: Fouling reversibility and the role of applied pressure



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## ABSTRACT

In this study, we systematically investigated the propensity and reversibility of combined organic–colloidal fouling in forward osmosis (FO) under various solution chemistries (pH and calcium ion concentrations) and applied hydraulic pressure on the feed side. Alginate, silica colloids, and their mixture (i.e., combined organic–colloidal) were used as model foulants. Our findings demonstrate that combined organic–colloidal foulants caused more rapid flux decline than the individual foulants due to the synergistic effect of alginate and silica colloids. As a result, much lower flux recovery was achieved by physical cleaning induced by increasing the cross-flow rate, in contrast to single foulants of which the fouling layer was easily removed under all solution conditions. Interestingly, less flux decline was observed at neutral pH for combined fouling, while acidic conditions were favorable for alginate fouling and basic solutions caused more silica fouling, thereby providing clear evidence for the combined fouling effect. It was also found that calcium ions enhanced water flux decline and induced the formation of less reversible combined organic–colloidal fouling layers. Lastly, the role of applied hydraulic pressure on the feed side in FO was examined to elucidate the mechanism of fouling layer formation, fouling reversibility, and water flux recovery. Higher fouling propensity and lower fouling reversibility of combined organic–colloidal fouling were observed in the presence of applied hydraulic pressure on the feed side. This observation suggests that the lower fouling propensity and greater fouling reversibility in FO compared to reverse osmosis (RO), are attributable to unpressurized operating conditions in FO.

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## 1. Introduction

Water shortages and depletion of fresh water supplies, likely exacerbated by climate change, place a great demand for alternative water resources. The urgent need for securing high quality water supply from unconventional sources, such as seawater and wastewater, has resulted in the emergence of advanced membrane technologies. Compared with thermal driven desalination, membrane processes based on reverse osmosis (RO) technology are much more energy efficient, with a global market of RO desalination increasing by more than 10% annually [1]. Despite the major advancements in RO desalination technology, its efficiency and sustainable operation are hampered by the relatively high energy consumption [2]. To reduce energy demand and costs, innovative system designs, such as multi-stage seawater reverse osmosis (SWRO) [2–5], energy saving processes that can utilize low grade

heat [4,6], and new membrane processes, including forward osmosis (FO) [1,7,8], have been suggested.

Forward osmosis has recently attracted heightened attention due to the wide range of prospective applications in desalination and wastewater reuse. Unlike the hydraulic pressure-driven RO process, water flux in FO is driven by osmosis due to a concentration difference between the feed water and a concentrated draw solution [7,9]. The apparent advantages of FO technology stem from its operation without applied hydraulic pressure, which has the potential for lower capital and operation costs [7,9–12]. The lack of applied hydraulic pressure on the feed side in FO has also been proposed as beneficial for fouling control compared to pressure-driven membrane processes [7,9,12–16].

Membrane fouling is a major obstruction in many membrane applications, which compromises process efficiency and increases operation and maintenance costs [17–20]. Several research efforts have been made to clarify the FO fouling mechanisms for expanding FO applications. Organic fouling behavior in FO was studied and compared to that in RO, demonstrating that under identical physicochemical conditions, more flux decline was generally

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observed in FO compared to RO due to cake-enhanced osmotic pressure that was accelerated by reverse permeation of draw solution [14]. However, higher operating cross-flow velocity was able to restore the initial flux by removing FO fouling layer, while such improvement was not observed in RO because of the compact fouling layer [14]. A more recent study investigated colloidal fouling in FO focusing on the role of reverse salt diffusion [21]. Similar to organic fouling, colloidal fouling induced elevated salt concentration near the membrane surface at feed side, primarily due to reverse salt flux from the draw to the feed side, which resulted in significant reduced water flux. However, in the absence of particle destabilization by increased salt concentration, the colloidal fouling layer was reversible upon increasing the cross-flow rate [21].

Although significant efforts have been made to understand fouling phenomena in FO, fundamental studies on fouling reversibility under various feed chemical compositions are rather limited. Particularly, there are no fundamental studies which investigate the role of hydraulic pressure in fouling, which is one of the most important factors discriminating fouling mechanisms between FO and RO. The main objective of this study was to evaluate the fouling propensity and reversibility of FO with a mixture of organic and colloidal foulants. Specifically, the mechanisms of combined organic and colloidal fouling were elucidated by examining the effects of individual foulants as well as synergistic effects with the combined organic and colloidal foulants. We have placed special emphasis on the role of osmotic pressure in fouling and fouling reversibility in FO compared to the role of hydraulic pressure in RO, which has been an issue of great interest in the literature. By designing fouling experiments in FO with increasing applied pressures on the feed side while maintaining the initial water flux at a fixed value, we clearly showed that osmotic pressure driving force results in more reversible fouling layers compared to hydraulic pressure driving force.

## 2. Materials and methods

### 2.1. FO membrane

A commercially available cellulose triacetate (CA) membrane from Hydration Technology Innovations (Albany, OR, USA) was used in this study. Membrane samples were stored in deionized (DI) water at 4 °C and then soaked in DI water at room temperature for 24 h before each experiment. For each fouling test, a fresh membrane sample was cut and placed into a bench-scale cross-flow membrane test cell in the typical FO mode (i.e., active layer faces feed solution). The total thickness of the membrane is approximately 50  $\mu\text{m}$  and other membrane properties are described elsewhere [21].

### 2.2. Organic and colloidal foulants

Two different types of model foulants, organic (alginate macromolecules) and inorganic (silica colloids), were used to study combined organic–colloidal fouling in FO. The model foulants represent polysaccharides and suspended colloidal matter, respectively, which are commonly found in real feed waters. We have used sodium alginate (12–80 kDa) as a model hydrophilic organic foulant (Sigma-Aldrich, St. Louis, MO, USA). For fouling experiments, alginate was added to the feed solution from a concentrated (2 g/L) sodium alginate stock solution. The concentration of alginate in the fouling experiments was 100 mg/L. Silica ( $\text{SiO}_2$ ) colloids were selected as a model particulate foulant. Silica colloids were added to the feed solution from a stock suspension (ST-ZL, Nissan Chemical Industries, Ltd., Japan). For fouling experiments,

the concentration of silica colloids in the feed solution was 1 g/L. According to information from the manufacturer, the approximate size of the colloidal particles is 100 nm. The particles were prepared as a stable concentrated aqueous suspension at an alkaline pH of 8.5–9.5. Colloidal particles of the required concentration were dispersed in deionized water (DI) with conductivity of less than 1  $\mu\text{S}/\text{cm}$ . Prior to use, the stock suspension was stirred vigorously for 10 min for uniform dispersion of the colloidal particles.

The model foulants were examined individually, and then, their mixture at a certain ratio was investigated in fouling experiments under different feed solution chemistries (i.e., pH and calcium ion concentration). For meaningful comparison of the fouling runs, similar hydrodynamic conditions (i.e., 7  $\mu\text{m}/\text{s}$  initial water flux and 8.61 cm/s cross-flow velocity) and feed solution chemistries were employed for all experiments.

### 2.3. FO lab-scale test unit

Experiments were performed in two different lab-scale membrane test units to investigate the fouling propensity and reversibility in FO. The first FO membrane test unit consisted of a monomer-cast (MC) nylon flow cell, two reservoirs for feed and draw solutions, two variable speed gear pumps (Cole-Parmer, Vernon Hills, IL) for circulation of each solution, a digital scale for measurement of permeate water mass, two customized salt-resistant Teflon flow meters, and a thermostatic bath. The various components of the unit were connected with Teflon tubing. A schematic diagram and detailed description of the FO system are given elsewhere [22].

The second FO unit was specially designed for application of hydraulic pressure on the membrane feed side (Fig. 1). An identical variable speed gear pump was used for the circulation of the draw solution as a typical FO system, but a high pressure pump replaced the gear pump on the feed side to apply hydraulic pressure. To prevent membrane breakage by the applied pressure, several layers of tailored permeate carriers obtained from commercial flat-sheet RO membrane modules (Hydranautics Inc., U.S.) were inserted as spacers on both sides of cell channel. These spacers are porous enough to allow flow of the solutions, and firm enough to bear the direct impact of pressure on the membrane surface. The dimensions of the feed and draw solution channels in both FO units were 77 mm long, 26 mm wide, and 3 mm deep.

### 2.4. Fouling tests

Fouling and cleaning experiments were conducted with the bench-scale membrane cross-flow units described above. The first

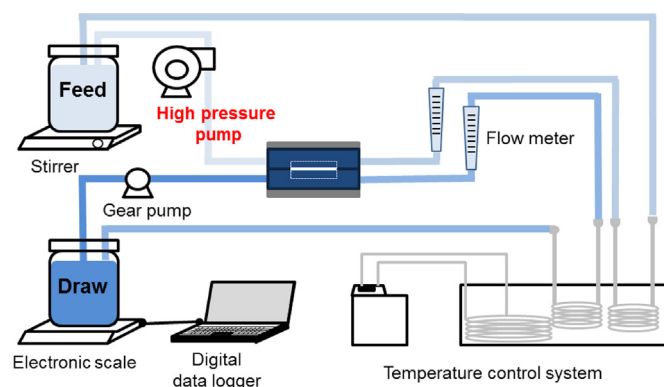


Fig. 1. Schematic description of the cross-flow FO membrane test unit specially designed for applying hydraulic pressure on the feed side of the membrane in order to investigate the impact of applied hydraulic pressure on fouling in FO.

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