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# Organic fouling in forward osmosis membranes: The role of feed solution chemistry and membrane structural properties



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## ABSTRACT

This study was aimed at investigating the effect solution total ionic strength, divalent ion concentration ( $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) and membrane structural properties on the fouling propensity of alginate. The fouling reversibility of the resulting fouling layer was also determined through the use of ultrapure water and salt solutions as cleaning agents. Sodium alginate was used as a model foulant to represent extracellular polymeric substance (EPS) that are present in seawater and wastewater. In order to understand the influence of all possible interactions the membrane was tested using solutions of two ionic strengths (0.1 M and 0.5 M) with varying concentrations of calcium and magnesium ions. Experimental results suggested that membrane orientation had an impact on fouling behavior since the membrane fouled more easily when operated in PRO mode than in FO mode. There was severe permeate flux decline in PRO mode mainly due to the calcium–alginate complexes blocking the pores in the support layer. The interfacial free energies obtained from advanced contact angle measurements correlated strongly with the rates of membrane fouling and further predicted the membrane fouling trends. Initial adhesion of alginate particles on the membrane surface was dictated by the chemical interactions between the membrane and alginate. It was also found that once the membrane surface is covered with the foulant, fouling becomes less sensitive to the changes in hydrodynamic conditions (permeation drag and cross-flow velocity) but rather depends on the foulant–foulant interactions. The resulting fouling layer was easily removed by shear force resulting from increased cross-flow velocity during membrane cleaning.

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## 1. Introduction

High pressure membrane filtration processes such as nanofiltration (NF) and reverse osmosis (RO) are presently the most widely applied processes for seawater desalination and wastewater reclamation to augment potable water supplies. These processes have proved to be capable of rejecting most dissolved constituents, including trace organic chemicals [1,2]. Provided that adequate pre-treatment is implemented, these processes can produce high quality potable water from reclaimed water, seawater, or from surface or ground water impacted by discharge from wastewater treatment plants [3]. However, since they require high hydraulic pressure to drive water across

the membrane, they are regarded as energy intensive and costly, especially with the current increasing energy prices. In addition to the high energy demand, the efficiency of NF and RO membranes is also thwarted by the inherent problem of membrane fouling that usually requires intensive chemical cleanings which can further escalate the treatment costs and reduce membrane lifetime [4–6].

Therefore it became imperative to investigate other possible alternative technologies for seawater desalination and wastewater reclamation, amongst which forward osmosis (FO) membrane filtration has become one of the most promising in the past years. Indeed, forward osmosis has over the past years received worldwide attention as promising alternative technology due to its claimed high resistance to fouling and wide range of pollutant rejection [7–10]. The distinguishing feature for FO when compared to RO is that FO utilizes the natural osmotic pressure gradient generated by a concentrated draw solution as the driving force for the net movement of water molecules through a semi-permeable membrane. The concentrated draw solution is diluted by water permeating from the feed water to

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desalination, and the resulting diluted draw solution can then be re-concentrated to recycle the draw solutes as well as to produce purified water. When compared to pressure-driven membrane processes, FO is claimed to possess a number of potential advantages including high rejection of a wide range of contaminants, low energy consumption and brine discharge, simple configurations and equipment as well as low membrane fouling propensity [11–13]. Nevertheless, similar to other membrane processes, the efficiency of FO processes is still thwarted by the enduring problem of membrane fouling and despite the numerous publications on the topic, there is still a lack of full systematic and mechanistic understanding of the fouling behavior for FO processes.

Membrane fouling in FO membrane filtration processes is hugely impacted by various factors ranging from hydrodynamic operating conditions to physical and chemical interactions between foulants and the membrane. Previous studies have confirmed lower fouling propensity for FO membranes compared to RO, mainly due to the lack of hydraulic pressure in driving water across the semi-permeable membrane [14–16]. Mi and Elimelech [17] investigated the role of calcium binding, initial permeate flux and membrane orientation in organic fouling of forward osmosis membranes using alginate, bovine serum albumin (BSA) and humic acid (HA). It was reported that calcium binding, permeation drag and hydrodynamic shear force were the major factors governing the development of a fouling layer on the membrane surface. They also performed subsequent membrane cleaning to determine the fouling reversibility behavior, whereby the results illustrated that alginate fouling in FO membrane processes is almost fully reversible by simple physical cleaning for a relatively short time [18].

Hong et al. [19] reported that the key mechanism of flux decline in FO is rather accelerated cake-enhanced osmotic pressure (CEOP) due to reverse salt diffusion from draw solution to the feed than an increase in fouling layer resistance, after conducting fouling experiments with alginate, humic acid and BSA, as well as silica colloids (SiO<sub>2</sub>). A more recent study by Yong Ng and Parid [20], focused on the impact of lower organic loads (10, 30, 50 ppm) in secondary effluents with calcium inclusion on the fouling characteristics of FO membranes both in the FO and PRO modes. In their work, they demonstrated that the FO mode (active layer facing feed water) had lower fouling compared to the PRO mode (active layer facing draw solution), which was also seen by other authors [17–22]. This was attributed to the denser, smoother and tighter structure of the membrane active layer which prevented the adhesion and accumulation of foulants on the membrane surface, while the porous support layer, being a looser structure, allowed the accumulation and deposition of the foulants on its surface and inside the membrane, by the mechanisms of direct interception and subsequent pore plugging. It was also found that the presence of 5 mM of Ca<sup>2+</sup> in the feed solution containing NOM, humic acid and biopolymers caused severe membrane fouling in the PRO configuration (about 85% water flux reduction in 20 h), due to the binding of calcium to carboxylic groups in natural organic matter result in large NOM clusters that settled easier on the loose support layer.

The internal concentration polarization (ICP) phenomenon has also been reported in several studies as another contributing factor to flux decline in FO membrane processes due to the reduction of the osmotic drive force. Tang et al. [21] investigated the coupled effect of organic fouling and internal concentration polarization (ICP) on FO flux behavior. They reported that ICP played a dominant role on FO flux behavior at higher draw solution concentrations and/or greater membrane fluxes due to their exponential dependence of ICP on flux levels. They observed that the active layer-facing draw solution (PRO) configuration was highly prone to flux reduction due to combined effects of internal clogging of the support layer and the resulting enhanced ICP in the support layer; the latter being caused by reduced porosity and reduced mass transfer coefficient of the support layer. Similar observations were made by Zhang et al. [22], who studied the

complex fouling phenomenon of FO membranes in a FO membrane bioreactor, where they discovered that a biofouling layer covered the substrate surface of the FO membrane with a combined structure of bacterial clusters and extracellular polymeric substances (EPS), which contributed to a massive drop in membrane support layer mass transfer coefficient.

In addition to solution chemistry and hydrodynamic conditions, membrane surface properties also greatly influence flux decline in membrane based filtration systems, since they impact membrane surface related processes such as wetting, adhesion and adsorption [23]. Foulant rejection potential is an another process that can be influenced by the membrane surface due to its affinity towards certain foulants. The magnitude of the chemical interactions between the membrane surface and foulant can be better predicted from determining the membrane surface free energy, which is generally explained by the extended Derjaguin–Landau–Verwey–Overbeek (XDLVO) theory. The XDLVO describes surface free energy as the sum of the apolar Lifshitz–van der Waals (LW) component and the polar or Lewis acid–base (AB) component [24]. Traditionally, this theory has been used to characterize membrane–colloid/aggregate interactions. Childress et al. [25] evaluated the surface energetics of three RO membranes and silica colloids using the XDLVO approach. The membranes were found to have low surface energies compared to the colloid and the interaction energy between the membranes and colloids was primarily dictated by the surface energies of the colloids.

In another work Lee and co-workers [26] used the XDLVO approach to describe NOM fouling in terms of interaction forces between NOM macromolecules and a membrane surface. They reported that acid–base (AB) interaction forces were dominant at short distance (< 5 nm) and determined the short-range foulant–membrane interactions. The obtained negative cohesion energy values predicted strong attractive hydrophobic interactions amongst the NOM macromolecules. While the similar negative adhesion energy values predicted strong attractive interactions between the NOM macromolecules and the membrane surface.

Jin et al. [27] examined the impacts of major ions in seawater on the acid–base properties of seawater RO membranes and alginic acid. They also evaluated correlations between (alginate–membrane and alginate–alginate) acid–base free energies and fouling. It was revealed that in membrane processes, initial organic adhesion is mediated by interfacial interactions between the organic molecules and clean membrane surface. After the membrane surface is covered by a layer of absorbed organics, subsequent organic adhesion is controlled by interfacial interactions between newly deposited and already deposited organics.

These studies have given more insight onto the cryptic mechanisms governing RO and FO membrane fouling by various foulants. However, since forward osmosis processes are aimed at purifying impaired water sources such as seawater more work needs to be done in elucidating the influence of the presence of cationic species on the foulant–membrane chemical interactions. It has been hypothesized that the acid–base properties of membranes and alginate are a function of the ionic composition of the aqueous media they occur in [27], therefore, developing a systematic understanding of FO membrane fouling behavior and the relation with foulant–membrane interactions in different ionic environments is of paramount importance.

The main focus of this work was at establishing a complete systematic understanding of the underlying fouling mechanisms of FO membranes by organic foulants at ionic concentrations as high as observed in seawater desalination processes. This was achieved by exposing the FO membrane to feed solutions of varying ionic concentrations of NaCl, CaCl<sub>2</sub> and MgCl<sub>2</sub>. Alginic acid (sodium alginate) was used as a model foulant to represent extracellular polymeric substance (EPS) found in both seawater and wastewater. The

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