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Fouling of reverse osmosis membranes by hydrophilic organic matter: implications for water reuse

Sangyoun Lee, Wui Seng Ang, Menachem Elimelech*

Department of Chemical Engineering, Environmental Engineering Program, P.O. Box 208286, Yale University, New Haven, CT 06520-8286, USA

Tel. +1 (203) 432-2789; Fax +1 (203) 432-2881; email: menachem.elimelech@yale.edu

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Abstract

Effluent organic matter (EfOM) is suspected as a major cause of fouling of reverse osmosis (RO) membranes in advanced wastewater reclamation. Among the main constituents in EfOM, polysaccharides are the most ubiquitous. The influence of solution chemistry and hydrodynamics on RO membrane fouling with alginate — a model for polysaccharides in secondary wastewater effluent — was systematically investigated. Results of fouling runs with alginate demonstrate that RO membrane fouling increases with decreasing pH, increasing ionic strength, and addition of calcium ions. At fixed solution ionic strength and pH, the presence of divalent calcium ions, at concentrations typical of those found in secondary wastewater effluent, had a dramatic effect on membrane fouling. However, for similar concentrations of divalent magnesium ions, fouling was negligible. The severe fouling in the presence of calcium is attributed to the formation of a thick, dense alginate gel layer on the membrane surface via calcium-alginate complexation and crosslinking (bridging) of alginate macromolecules by calcium. In addition to solution chemistry, hydrodynamic operating conditions — initial permeate flux and crossflow velocity — were also shown to influence RO membrane fouling with alginate.

Keywords: Alginate; Polysaccharide; Wastewater reclamation; RO fouling; Divalent cations; Organic fouling

1. Introduction

In recent years, membrane processes have emerged as a viable technology for water and

wastewater treatment. A successfully implemented membrane technology can produce water of superior quality from a variety of source waters compared to conventional technologies. Of particular interest is the use of reverse osmosis (RO)

*Corresponding author.

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in advanced wastewater reclamation, using secondary wastewater effluent as source water.

A typical scheme for advanced wastewater reclamation involves the following steps: (i) biological wastewater treatment, (ii) pretreatment of the secondary effluent with microfiltration (MF) to remove suspended and colloidal matter, (iii) treatment of effluent with RO, and (iv) UV disinfection of product water. Pretreated secondary effluent, however, still contains considerable amount of organic macromolecules, designated as effluent organic matter (EfOM), which are problematic due to their potential contribution to organic fouling of RO membranes. Therefore, understanding the causes of RO membrane fouling by EfOM and developing strategies for fouling control are of paramount importance for successful application of RO technology in water reuse.

Effluent organic matter (EfOM) comprises a wide range of compounds, often measured as dissolved organic carbon (DOC), ranging from low to high molecular weight compounds. Examples of such compounds include polysaccharides, proteins, amino-sugars, nucleic acids, humic and fulvic acids, organic acids, and cell components [1,2]. Among these, polysaccharides are some of the most ubiquitous hydrophilic macromolecules in secondary effluent. Polysaccharides are produced during biological wastewater treatment processes and they are part of the so-called soluble microbial products (SMP) [2,3].

Alginate is an acidic polysaccharide produced by bacteria, microalgae, and macroalgae, and is typically made up of repeating manuronic and guluronic acids [4–6]. In the area of wastewater treatment employing biological treatment processes, numerous studies have investigated the physicochemical properties of alginate since it plays an important role in bioflocculation, and thus governs the efficiency of solid/liquid separation, settling, and dewatering [7–9]. However, no systematic studies are available that address the role of alginate in the fouling of RO membranes.

The main objective of this study is to systematically investigate the influence of feed water chemistry and hydrodynamic operating conditions on alginate fouling of RO membranes. Specifically, we investigate the effects of chemical (solution pH, ionic strength, and divalent cations) and physical (initial flux and crossflow velocity) factors on the alginate fouling mechanisms. Emphasis is placed on the influence of divalent cations (Mg^{2+} and Ca^{2+}), which are important ionic constituents of natural and wastewaters.

2. Materials and methods

2.1. Chemical reagents and model alginate

All chemicals were reagent grade and used without further purification. ACS grade NaCl, $CaCl_2 \cdot 2H_2O$, and $MgCl_2 \cdot 6H_2O$ salts, as well as trace metal HCl and NaOH, were obtained from Fisher (Pittsburgh, PA). Deionized (DI) water (Nanopure Infinity Ultrapure, Branstead, Dubuque, IA) was used for the preparation of all the stock solutions and for membrane fouling experiments.

A commercial alginic acid (Sigma-Aldrich, St. Louis, MO) extracted from brown algae was chosen as a model hydrophilic organic matter. The alginic acid was received in a powder form and the stock solution (2 g/l) was prepared by dissolving the alginic acid in DI water completely, followed by filtration with a 0.45- μm filter (Durapore, Millipore, Billerica, MA). Based on the manufacturer, the molecular weight of the alginate ranges from 12 to 80 kDa. The carboxylic acidity measured by potentiometric titration (794 Basic Titrino, Metrohm, Switzerland) was shown to be 10.5 meq/g Carbon (or 3.3 meq/g alginate); most carboxylic groups were deprotonated above pH 5.

2.2. RO membrane and crossflow test unit

The relatively well characterized thin-film composite LFC-1 (Hydranautics, Oceanside, CA) was used as a model RO membrane in the fouling experiments [10]. Average hydraulic resistance

and NaCl rejection were determined to be 9.16 (± 0.11) $\times 10^{13}$ m⁻¹ and 98.5 (± 0.3) %, respectively. The membrane has been reported to be negatively charged at solution chemistries typical to natural and waste waters, with an isoelectric point at about pH 4 [10].

A laboratory-scale crossflow membrane test unit was used for the fouling experiments. The test unit is comprised of a membrane cell, high pressure pump (Hydra-cell pump, Wanner Engineering, Minneapolis, MN), back-pressure regulator (US Paraplate, Auburn, CA), bypass valve (Swagelok, Solon, OH), feed water reservoir (Nalgene, Rochester, NY), magnetic stirrer (Cimarec 2, Branstead/Thermolyne, Dubuque, IA), digital flow meter (Optiflow 1000, Humonics, Folsom, CA), floating disc flow meter (Blue-White, Huntington Beach, CA), and temperature controller (RET-111, NESLAB, Newington, NH). The rectangular plate-and-frame membrane cell has dimensions of 7.7 cm length, 2.6 cm width, and 0.3 cm height. The system was operated in a closed loop mode with both permeate and retentate being recirculated into the feed water reservoir. The back-pressure regulator in conjunction with the bypass valve at the entrance allowed fine control of trans-membrane pressure and retentate flow rate. Temperature was maintained constant during the fouling runs by a chiller with a stainless-steel coil submerged in the feed water reservoir. A digital flow meter interfaced with a PC was used to continuously monitor the permeate flux.

2.3. Fouling experiments

Alginate fouling experiments were performed with the crossflow test unit described in the previous section. The alginate concentration and feed solution chemistry (i.e., pH, ionic strength, and divalent cation concentration) were varied for each set of fouling experiments. The hydrodynamic operating conditions (i.e., initial flux and crossflow velocity) for each set of fouling experiments were also varied as needed. When investi-

gating the influence of feed solution chemistry on fouling, the initial flux and the crossflow velocity were fixed at 20 μ m/s (42 gfd) and 8.5 cm/s, respectively. Similarly, the alginate concentration and the feed solution chemistry were fixed (i.e., alginate concentration = 20 mg/l; pH 6.0; calcium concentration = 0.5 mM; and total ionic strength = 10 mM, by varying NaCl concentration) when investigating the influence of hydrodynamics on fouling.

The protocol developed for alginate fouling experiments is summarized in Table 1. The membrane was first compacted with DI water in the membrane test cell for 6 h. Then, the membrane was stabilized and equilibrated with alginate-free electrolyte solution (pH, ionic strength, and divalent cation concentration were the same as those used for the subsequent fouling run) for 6 h. The purpose of this step was to achieve a stable permeate flux and to yield reproducible results during the ensuing fouling runs. After attaining a stable permeate flux, the initial flux and the crossflow velocity were adjusted to the desired value by adjusting the trans-membrane pressure and retentate flow rate, respectively. Following the adjustments of initial flux and crossflow velocity, the performance of the membrane was evaluated in terms of permeate flux and salt rejection. These performance data served as a baseline for the subsequent fouling experiments. Fouling was

Table 1
Experimental protocol for RO fouling tests

Time (h)	Description
0–6	Compaction with DI water
6–12	Adjustment of solution chemistry (pH and ionic strength) and stabilization/equilibration
12–18	Adjustment of hydrodynamic conditions (initial flux and crossflow velocity) and baseline performance
18–36	Addition of alginate and initiation of fouling experiment

initiated by adding an alginate stock solution to the feed water to achieve the desired alginate concentration (20 or 50 mg/l). The change in permeate flux was monitored continuously throughout the fouling tests and the corresponding flux decline curves were used to assess the alginate fouling behavior under the various chemical and physical conditions employed during each fouling test.

3. Results and discussion

3.1. Influence of solution chemistry on fouling

3.1.1. Solution pH

The influence of solution pH on alginate fouling is shown in Fig. 1. A more significant decline in permeate flux is observed at pH 3 compared to pH 6 and 9. This behavior is attributed to the absence of electrostatic repulsion among the alginate macromolecules and between alginate and the membrane surface at pH 3. At pH 3, the alginate macromolecules are protonated and thus are neutrally charged. Furthermore, at this pH, the surface charge of the membrane is neutral or even slightly positively charged as the isoelectric point of the

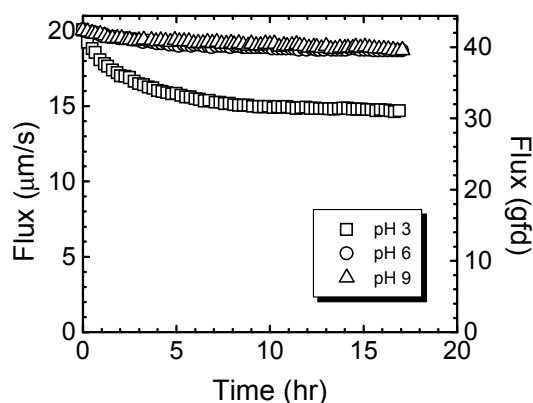


Fig. 1. Effect of solution pH on alginate fouling. The pH of the feed solution is adjusted by adding 0.1 M HCl or NaOH stock solutions. Experimental conditions: alginate concentration = 20 mg/l, total ionic strength = 30 mM NaCl, initial permeate flux = 20 $\mu\text{m/s}$ (42 gfd), crossflow velocity = 8.5 cm/s, and temperature = 20.0 \pm 0.5 $^{\circ}\text{C}$.

membrane is at pH 4. These conditions promote alginate deposition and accumulation on the membrane surface which result in fouling.

The very small flux decline at pH 6 and 9 is because all the alginate carboxylic groups are already deprotonated at pH 5, and the membrane is negatively charged at both pH 6 and 9. Under these conditions, alginate accumulation on the membrane surface is not substantial. In addition, the alginate macromolecules have a stretched and less compact configuration above pH 5, due to increased electrostatic repulsion between negatively charged neighboring carboxyl groups, and thus form a sparser fouling layer. This loose alginate layer does not provide significant hydraulic resistance to permeate flow. Similar trends of increased flux decline with decreasing pH for charged natural organic matter (NOM) macromolecules were observed by Hong and Elimelech [11].

3.1.2. Ionic strength

To investigate the influence of ionic strength on alginate fouling, fouling experiments were performed at four different ionic concentrations of an indifferent electrolyte — 10, 30, 50, and 100 mM NaCl. Fig. 2 shows clearly that alginate fouling becomes more visible as the ionic strength of the feed solution increases. At higher ionic strength, the charges of the alginate macromolecules and the membrane are reduced due to double layer compression or charge screening, leading to a decrease in electrostatic repulsion among alginate macromolecules and between the alginate and the membrane surface. As a result, alginate deposition onto the membrane surface increases and the alginate fouling layer becomes thicker. In addition, due to reduced interchain electrostatic repulsion at high ionic strength, alginate macromolecules become more coiled and form a more compact fouling layer. The resulting fouling layer provides an additional hydraulic resistance to permeate flow through the membrane and leads to visible flux decline. In contrast, at low ionic strength, the alginate fouling layer does not sig-

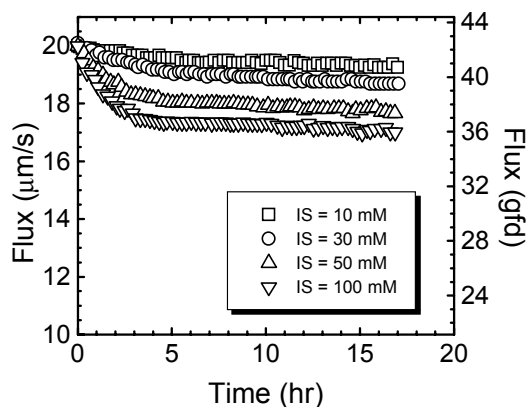


Fig. 2. Effect of ionic strength (IS) on alginate fouling. The total ionic strength of the feed solution is adjusted by varying NaCl concentration. Experimental conditions: alginate concentration = 20 mg/l, pH = 6.0 ± 0.1, initial permeate flux = 20 µm/s (42 gfd), crossflow velocity = 8.5 cm/s, and temperature = 20.0 ± 0.5 °C.

nificantly contribute to the increase in the total hydraulic resistance to permeate flow. Under these conditions, the strong electrostatic repulsion among alginate macromolecules and between alginate and the membrane surface prevents significant alginate accumulation on the membrane surface. At lower ionic strength, alginate

also forms a much looser fouling layer, since alginate macromolecules have a linear configuration. Such a loose fouling layer provides very little resistance to permeate flow and, hence, negligible flux decline.

3.1.3. Divalent cations

The influence of divalent cations (Mg^{2+} and Ca^{2+}) on alginate fouling is shown in Fig. 3. Fouling experiments were performed at two different divalent cation concentrations — 0.5 and 1.0 mM — and with no divalent cations. Total ionic strength was kept constant (10 mM) by varying the NaCl concentration. The results clearly demonstrate that permeate flux decreases dramatically in the presence of calcium ions, but not with magnesium ions. This behavior can be explained by the unique structure of the alginate fouling layer, formed through calcium-alginate complexation followed by calcium bridges between adjacent alginate macromolecules. This crosslinked alginate fouling layer formed in the presence of calcium produces a substantial resistance to permeate flow, leading to a severe flux decline. It is important to note that the severe flux decline due to alginate fouling in the presence of calcium results from

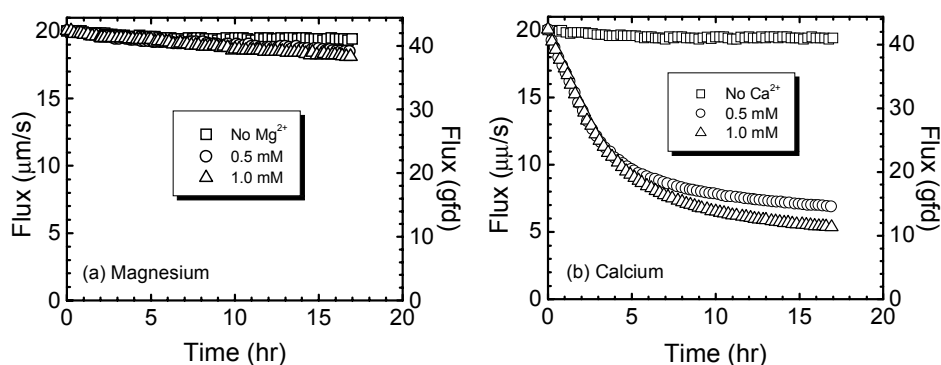


Fig. 3. Effect of divalent cation concentration on alginate fouling: (a) magnesium and (b) calcium. The total ionic strength of the feed solution is fixed at 10 mM by varying NaCl concentration (i.e., feed solution with no Mg^{2+} or Ca^{2+} contains 10 mM NaCl, feed solution with 0.5 mM Mg^{2+} or Ca^{2+} contains 8.5 mM NaCl, and feed solution with 1.0 mM Mg^{2+} or Ca^{2+} contains 7.0 mM NaCl). Experimental conditions: alginate concentration = 20 mg/l, pH = 6.0 ± 0.1, initial permeate flux = 20 µm/s (42 gfd), crossflow velocity = 8.5 cm/s, and temperature = 20.0 ± 0.5 °C.

foulant–foulant interaction; that is, interaction between alginate in the bulk phase and alginate deposited onto the membrane surface. Therefore, this specific interaction of alginate with calcium, but not with magnesium ions, and the subsequent alginate gel formation are the major cause for the severe flux decline in the presence of Ca^{2+} compared to Mg^{2+} .

Calcium induced gelation of alginate has been widely explained by a co-operative model, known as the ‘egg-box’ model [12]. Based on this model, divalent cations (such as calcium) bind preferentially to alginate in a highly cooperative manner and form bridges between adjacent alginate macromolecules, leading to the ‘egg-box’ shaped gel network. Alginate gel formation in the presence of calcium has been reported by several researchers. Davis and co-workers [6] reported an enhanced selectivity of alginates, extracted from several species of *Sargassum* seaweed, for calcium and cadmium relative to smaller divalent cations, such as magnesium, leading to gel network formation. Bruus and co-workers [7] demonstrated that physical properties of biological flocs (e.g., specific resistance to filtration) formed with alginate in the presence of calcium are improved significantly compared to those in the absence of calcium, thus inferring that calcium induced alginate gel formation is important in the bioflocculation process. They also observed that the properties of biological flocs formed with alginate in the presence of magnesium were severely deteriorated compared to the case with calcium, thus concluding the biopolymer had greater affinity for Ca^{2+} than Mg^{2+} . The influence of Ca^{2+} and Mg^{2+} on humic acid fouling of NF membranes has been investigated by Li and Elimelech [13]. Similar to our results, it was shown that permeate flux declined much more severely with calcium compared to magnesium. It was also demonstrated that intermolecular adhesion forces between humic molecules and the fouled membrane increased significantly with calcium compared to magnesium. We suggest that the fouling mechanisms with humic

acid reported by Li and Elimelech [13] are applicable to our results with alginate. In fact, we confirmed that a thick alginate gel layer was formed on the membrane surface at the end of the fouling runs performed in the presence of calcium; however, no alginate gel layer was visible when the fouling run was carried out in the presence of magnesium.

Fig. 4 indicates that the initial fouling rate increased with increasing alginate concentration (from 20 mg/l to 50 mg/l) at fixed divalent cation concentration, especially with calcium ions. The behavior is comparable to the results shown in Fig. 3b (for calcium). In Fig. 3b, the initial fouling rates for 0.5 and 1.0 mM calcium at a fixed alginate concentration were almost the same. Hence, it can be inferred that the initial interaction between alginate and the membrane is mainly controlled by alginate concentration, whereas interaction between alginate in bulk phase and alginate in the fouling layer is mostly governed by calcium ion concentration. Within relation to the ‘egg-box’ model, the former interaction can be related to the binding between calcium and alginate and the latter interaction to the bridging between adjacent alginate macromolecules.

3.2. Influence of hydrodynamics on fouling

3.2.1. Initial flux

The influence of initial flux (or trans-membrane pressure) on alginate fouling is presented in Fig. 5. The flux decline behavior is plotted in three ways — flux vs. time (Fig. 5a), normalized flux vs. time (Fig. 5b), and normalized flux vs. cumulative permeate volume (Fig. 5c). The latter presentation of flux vs. cumulative volume, rather than time, is more appropriate when comparing fouling runs performed at different initial fluxes [11,14]. All fouling experiments were performed at the same alginate concentration (20 mg/l), solution chemistry (0.5 mM calcium, total ionic strength of 10 mM, and pH 6), and crossflow velocity (8.5 cm/s). A greater flux decline is observed

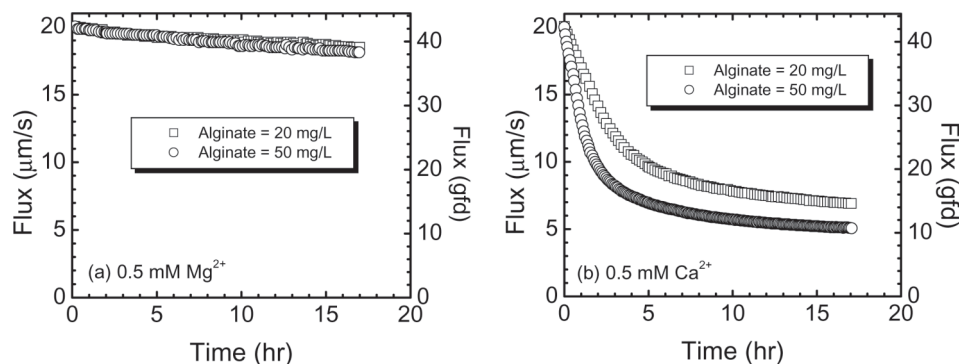


Fig. 4. Effect of organic foulant concentration on alginate fouling in the presence of divalent cations: (a) magnesium and (b) calcium. Experimental conditions: total ionic strength = 10 mM (0.5 mM Mg^{2+} plus 8.5 mM NaCl or 0.5 mM Ca^{2+} plus 8.5 mM NaCl), pH = 6.0 ± 0.1 , initial permeate flux = $20 \mu\text{m/s}$ (42 gfd), crossflow velocity = 8.5 cm/s, and temperature = $20.0 \pm 0.5 \text{ }^\circ\text{C}$.

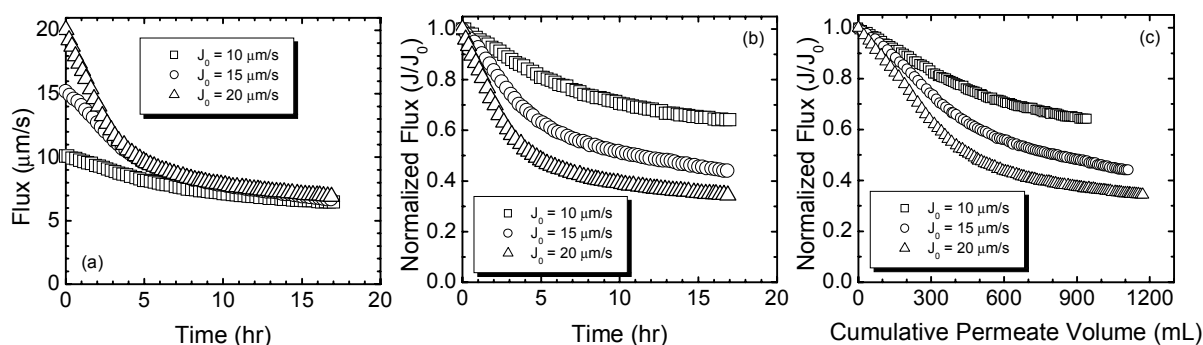


Fig. 5. Effect of initial flux (J_0) on alginate fouling: (a) permeate flux vs. filtration time, (b) normalized flux vs. filtration time, and (c) normalized flux vs. cumulative permeate volume. Experimental conditions: alginate concentration = 20 mg/l, total ionic strength 10 mM (0.5 mM Ca^{2+} plus 8.5 mM NaCl), pH = 6.0 ± 0.1 , crossflow velocity = 8.5 cm/s, and temperature = $20.0 \pm 0.5 \text{ }^\circ\text{C}$.

for higher initial flux, thus confirming our previous finding regarding the important role of the trans-membrane pressure in membrane fouling [11,14,15]. The greater flux decline at higher initial flux, even when flux is plotted against cumulative volume (Fig. 5c), is also related to the compressibility of the fouling layer. At higher initial flux (or trans-membrane pressure), the alginate fouling layer becomes more compressed, leading to an increased hydraulic resistance to permeate flow. The compressibility of organic fouling layers has been studied extensively for various organic

foulants, such as natural organic matter [11,14] and proteins [16–18]. These studies reported increased specific resistance of fouling layers with increasing trans-membrane pressure.

3.2.2. Crossflow velocity

The influence of crossflow velocity (or shear rate) on alginate fouling is illustrated in Fig. 6. The results clearly show that alginate fouling is lessened by increasing the crossflow velocity. The effect of crossflow velocity on alginate fouling is

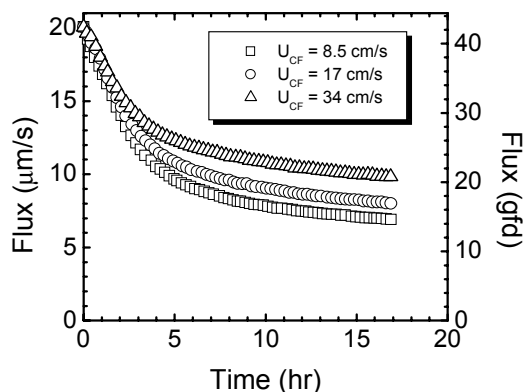


Fig. 6. Effect of crossflow velocity (U_{CF}) on alginate fouling. Experimental conditions: alginate concentration = 20 mg/l, total ionic strength = 10 mM (0.5 mM Ca^{2+} plus 8.5 mM NaCl), pH = 6.0 ± 0.1 , initial permeate flux = 20 mm/s (42 gfd), and temperature = 20.0 ± 0.5 °C.

attributed to the increase in shear rate and the resulting reduction in alginate and calcium concentrations at the membrane surface. It is noticeable that the initial fouling rate (up to 3 h of filtration) is similar for all flux decline curves irrespective of the crossflow velocity. Similar lack of dependence of fouling on crossflow velocity at the initial stages of filtration has been reported in for NOM [14] and colloidal [19] fouling.

4. Summary

The chemical composition of feed water greatly influences alginate fouling of RO membranes. Alginate fouling of RO membranes is accelerated at lower pH, higher ionic strength, and in the presence of calcium ions. Of these chemical factors, the presence of calcium ions has a much greater effect on fouling. The severe alginate fouling in the presence of calcium results from calcium induced alginate gel formation at the membrane surface. Compared to the severe fouling with calcium, no fouling is observed in the presence of magnesium. This behavior is attributed to the lack of alginate gel formation in presence of magnesium. In addition to chemical factors, hydrody-

namic conditions such as initial permeate flux and crossflow velocity also play an important role in alginate fouling. Increasing initial flux results in higher rate of alginate fouling, whereas increasing crossflow velocity lessens the extent of fouling. Thus, optimization of hydrodynamic conditions can be useful for organic fouling minimization.

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