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# Interaction energy evaluation of the role of solution chemistry and organic foulant composition on polysaccharide fouling of microfiltration membrane bioreactors

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## HIGHLIGHTS

- Effect of feed solution on polysaccharide fouling of MF membrane was studied.
- Polysaccharide fouling of MF membrane was evaluated by XDLVO theory.
- Measured interaction energies were related to fouling rate of MF membrane.
- Greater attraction and lower repulsion energies induced higher MF membrane fouling.

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## ABSTRACT

Microfiltration membrane bioreactors (MF MBRs) have been extensively applied in wastewater treatment. Polysaccharide has been known to contribute significantly to organic fouling of MF membranes. In this study, the influences of ionic strength, divalent cations ( $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) and organic foulant composition on polysaccharide fouling of MF membranes were investigated. Interfacial interaction parameters analysis showed that the cohesion and adhesion energies of polysaccharide increased with higher ionic strength, presence of divalent cations, and higher mass ratio of polysaccharide to protein. Fouling experiments indicated that MF membrane fouling by polysaccharide was also enhanced with increasing ionic strength, divalent cations addition, and higher polysaccharide concentration. Measured interaction energies confirmed the trends of the fouling profiles. Since the MF membrane surfaces were not completely coated with polysaccharide and some of the clean membrane remained exposed after fouling, it was consistently shown that feed solutions that induced higher fouling rates were associated with greater attractive and lower repulsive interaction energies among the polysaccharide molecules and between polysaccharide molecules and clean MF membrane.

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## 1. Introduction

Microfiltration membrane bioreactors (MF MBRs) have been actively employed for municipal and industrial wastewater treatment applications (Arabi and Nakhla, 2010). The obstacle to more widespread use of membrane technology is membrane fouling, which decreased permeability and added operational cost (Bouhabila et al., 2001; Tian et al., 2011). Polysaccharide was assumed to be one of the major membrane foulants (Drews, 2010). Moreover, there

was a linear relationship between fouling rate and polysaccharide concentration in the sludge supernatant (Rosenberger et al., 2006). Therefore, understanding polysaccharide fouling is critical for successful and widespread application of MF membrane in wastewater treatment.

Polysaccharide fouling is a complicated process due to the complex interaction between polysaccharide and membrane and between polysaccharide molecules. It has been reported that for reverse osmosis (RO) membrane, since monolayer coverage on the RO membrane surface through foulant–membrane interactions was attained in a very short period of time, the rate and extent of organic fouling were determined by the foulant–foulant interactions (Lee and Elimelech, 2006). Thus the analysis of foulant–foulant cohesion could provide molecular-level understanding the

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organic fouling mechanisms of RO membrane. However, it is worthwhile to note that the fouling behavior of MF membranes is likely to be very different from that of RO membranes. Pore blocking has been reported as an important fouling mechanism for porous MF membranes but is unlikely to be important for non-porous RO membranes (Ang and Elimelech, 2007; Wang and Tarabara, 2008). The fouling mechanisms of MF membranes have been attributed to membrane pore blocking and/or pore constriction during initial fouling, followed by cake formation on the membrane surface during long-term fouling (Güell and Davis, 1996). The formation of membrane fouling mainly consisted of two processes: first the foulants adsorbed to the clean membrane, and then the foulants adhered to the foulants on the membrane. Since a fraction of organic foulants could pass in MF membrane pores (Liang et al., 2007), the membrane surfaces were not completely coated with organic foulants and some of the clean membrane remained exposed after fouling in a period of time (Subramani et al., 2009). Hence, the polysaccharide fouling process of MF membrane including cake buildup and pore plugging may be controlled by the combined interaction energies among polysaccharide molecules and between polysaccharide molecules and MF membrane. During the MF operation, periodic membrane backwash and chemical cleaning were utilized to recover the membrane from “reversible” fouling (typically due to cake formation) and “irreversible” fouling (typically due to pore plugging), respectively. Furthermore, characterization of the physicochemical interactions between polysaccharide and MF membrane and between polysaccharide molecules would be helpful to increase the membrane cleaning efficiency.

Thus, this study focused on interactions between polysaccharide and membrane surface and between polysaccharide molecules, which were important stages of the much more complex overall process of MF membrane fouling. These interactions could be interpreted through extended Derjaguin–Landau–Verwey–Overbeek (XDLVO) models (Kang et al., 2004; Subramani and Hoek, 2008; Wang et al., 2005). Sodium alginate (SA) has been used as a model foulant to study organic fouling processes (Lee and Elimelech, 2006). Moreover, the ionic strength, divalent cations, and organic composition played important roles in adhesion force between SA molecules in bulk solution and SA molecules on the RO membrane (Lee and Elimelech, 2006). Nevertheless, the influence of solution characteristics on MF membrane fouling and interaction energies among SA molecules and between the SA molecules and MF membrane had been scarcely assessed. In previous study (Childress and Elimelech, 1996; Shim et al., 2002), they observed that the membrane showed different characteristics with different ionic strengths and ionic species. Meanwhile, membrane foulants readily adsorbed to the membrane surface and markedly influenced the membrane surface charge. This meant that the membrane surface charge would be continuously changed with the foulants adsorbed to the membrane. Therefore, the interaction between membrane surface and foulants was hardly determined due to the continuously changed membrane surface charge. It is widely known that the formation of a fouling layer during initial stages of membrane filtration leads to subsequent fouling layer development on the membrane surface. In this study, the initial membrane characteristics were used to determine the membrane fouling tendency.

The motivation of this research was, therefore, to contribute toward a better understanding the effect of feed solution characteristics on polysaccharide fouling of MF membrane based on XDLVO theory. Specifically, the changed fouling characteristics of polysaccharide with different ion strengths, divalent cations and feed foulant compositions were investigated by batch filtration experiments; the physicochemical interactions between polysaccharide and MF membrane and between polysaccharide molecules

were characterized through XDLVO models. The results obtained from the bench-scale fouling experiments and the corresponding interaction energies were used to elucidate the fouling mechanisms of MF membranes.

## 2. Materials and methods

### 2.1. Organic foulants

Sodium alginate (SA) and Bovine serum albumin (BSA) (Sigma-Aldrich, St. Louis, MO) were used as model organic foulants. Both BSA and SA have been used extensively to represent the more complex proteinaceous and heteropolysaccharide materials present in wastewater (de Kerchove and Elimelech, 2007; Kim and Hoek, 2007). Both organic foulants were received in powder form, and stock solutions (2 g/L SA and 10 g/L BSA) were prepared by dissolving the SA and BSA in deionized (DI) water. Mixing of the stock solutions was performed for over 24 h to ensure complete dissolution of foulants, followed by the filtration using a 0.45  $\mu\text{m}$  cellulose acetate membrane. The filtered stock solutions were stored in sterilized glass bottles at 4 °C.

Reagent grade chloride salts of sodium, calcium and magnesium were used to adjust ionic strength of feed water. Solution chemistries used for fouling experiments and interaction energy calculations included variations in electrolyte concentrations (NaCl) and divalent cations ( $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$ ). In all cases, identical solution chemistries were employed in both fouling experiments and interaction energy calculations. When investigating the effect of divalent cations ( $\text{Mg}^{2+}$  or  $\text{Ca}^{2+}$ ), the feed solution was amended to desired value by adding 0.1 M  $\text{MgCl}_2$  or  $\text{CaCl}_2$  stock solutions. Meanwhile, the total ionic strength was maintained constant by adjusting NaCl concentration. The organic foulant composition was varied as needed by adjusting the ratio of the SA to BSA concentration, while the total organic foulant concentration was kept constant. Measured physicochemical properties of organic foulants are listed in Table S1.

### 2.2. Microfiltration membranes

Polyvinylidene fluoride (PVDF) is advantageous over other membrane materials due to its high mechanical strength and excellent chemical resistance (Liu et al., 2011). This relatively well-characterized membrane has been extensively applied in microfiltration for general separation purposes (Liu et al., 2011). Therefore, the commercial PVDF (millipore) flat sheet membrane with a nominal pore size of 0.22  $\mu\text{m}$  was selected as a model membrane for the fouling experiments. The membrane characteristics are listed in Table S2. PVDF membranes were immersed in 75% (v/v) alcohol for ca. 2 h, ensuring the membranes were sufficiently wetted and degassed. Prior to use, all membranes were soaked in DI water for 24 h with several intermediate water changes to remove impurities or additives.

### 2.3. Filtration apparatus

To investigate the influence of feed solution characteristics on fouling, the fouling propensities of SA with different ionic strengths, divalent cations and organic foulant compositions were evaluated using a stirred dead-end cell (MSC300, Mosu Corp., China) at room temperature ( $20 \pm 1$  °C). The flat sheet PVDF membranes were employed for filtration with an effective membrane area of 19.62  $\text{cm}^2$ . Before each experiment, DI water was filtered through the membrane for 1 h to stabilize the filtration system. The filtration pressure was maintained constant at 10 kPa and the stirring speed in cell was set at 250 rpm throughout the

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