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Membrane fouling by sodium alginate in high salinity conditions to simulate biofouling during seawater desalination

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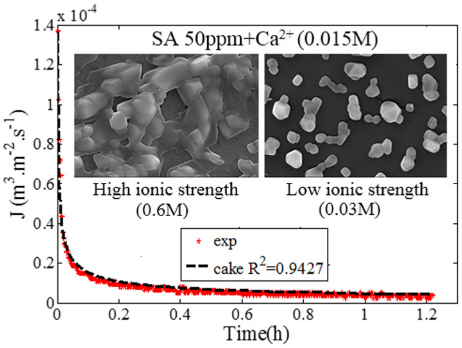
HIGHLIGHTS

- During sodium alginate microfiltration, fouling mechanisms depend on SA concentration.
- Na^+ (0.6 M) reduces membrane fouling during SA microfiltration.
- Ca^{2+} and Na^+ together leads to quick deposit formation regardless of SA concentration.
- In low ionic strength conditions (0.03 M) binding of SA by Ca^{2+} decreases.
- In high ionic strength conditions (0.6 M), SA is highly rejected by MF membrane.

GRAPHICAL ABSTRACT



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ABSTRACT

This study aims to better understand biofouling by algal organic matters (AOM) during seawater pretreatment by microfiltration (MF). To simulate AOM biofouling, sodium alginate (SA) solutions with three different concentrations (2, 20 and 50 ppm) were filtered in dead-end mode with MF membrane. A modelling approach with blocking laws was used to identify the fouling mechanisms behind flux decline with time. The effect of SA concentration and cations such as Na^+ (0.6 M) and Ca^{2+} (0.015 M) addition to SA solution on fouling mechanisms was studied. While for low SA concentration (2 ppm), fouling occurs within two phases: a pore constriction phase followed by cake formation phase, for high SA concentration (50 ppm), fouling occurs within only one phase controlled by cake formation. The addition of Na^+ (0.6 M) or Ca^{2+} (0.015 M) to SA solution mitigates membrane fouling, however, the addition of both cations enhances fouling by formation of dense cake layer on membrane.

1. Introduction

The high demand for water and the limitation of water resources makes it necessary to explore new alternatives such as wastewater treatment for reuse and seawater desalination. With regard to seawater desalination technologies, Reverse Osmosis (RO) membrane processes have been widely studied and have proven their efficiency (Amy et al., 2017; Aslam et al., 2017). Nevertheless, a filtration of raw seawater, rich in foulant material and especially colloidal matter using RO membrane accelerates membrane fouling and declines its performance (Jin et al., 2009; Miyoshi et al., 2016; Monnot et al., 2016). To improve desalination performance by RO membrane, a pretreatment to remove potential foulant materials prior to the RO process is necessary (Jeong et al., 2016; Monnot et al., 2016). Many conventional technologies based on chlorination, ozonation or coagulation-flocculation have been then largely studied (Jamaly et al., 2014). Even if many interests in seawater pretreatments are growing rapidly, the microfiltration (MF) and/or ultrafiltration (UF) as pretreatment to remove

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Nomenclature

A	membrane area (m^2)
C_{SA}	sodium alginate concentration ($kg.m^{-3}$)
e	membrane active layer thickness (m)
h	cake thickness (m)
J	permeate flux ($m^3.m^{-2}.s^{-1}$)
J_0	initial permeate flux ($m^3.m^{-2}.s^{-1}$)
K_{pc}	pore constriction parameter (m^{-1})
K_{cb}	complete blocking parameter (s^{-1})
K_{ib}	intermediate blocking parameter (m^{-1})
K_{cf}	cake formation parameter ($s.m^{-2}$)
m	cake mass (kg)
t	filtration time (s)
α	average specific cake resistance ($m.kg^{-1}$)
ΔP	transmembrane pressure (Pa)
μ	permeate viscosity (Pa.s)
ρ_c	cake density ($kg.m^{-3}$)
ρ_f	feed density ($kg.m^{-3}$)
σ	blocked area per unit volume of permeate ($m^2.m^{-3}$)

suspended solids as well as colloidal materials in seawater desalination still require more studies (Jamaly et al., 2014; Voutchkov, 2010; Xu et al., 2010, 2012).

Eutrophication occurring in aquatic system is responsible for algal bloom that leads to increased amount of algal organic matter (AOM). The occurrence of algal bloom in seawater has been considered as a main hurdle in deteriorating the whole performance in seawater treatment process. Being composed of high proportion of polysaccharides, (Pivokonsky et al., 2014; Qu et al., 2012) makes AOM potential membrane foulant which is able to decline membrane filtration efficiency (Babel and Takizawa, 2010; Peiris et al., 2013). Villacorte et al. (2015a, 2015b) showed that AOM such as transparent exopolymer particle, have a high fouling potential under both high salinity and low salinity conditions (Castaing et al., 2010). Moreover, they proved that the AOM produced by algal cells have higher fouling potential than the algal cells themselves. MF membrane fouling by AOM present in low salinity waters such as lakes or drinking waters reservoirs have been widely studied (Babel and Takizawa, 2010; Zhang et al., 2016, 2013). Nevertheless, MF membrane fouling by AOM in high salinity waters such as seawater has not been well studied.

To simulate AOM and polysaccharides, numerous studies have used sodium alginate (SA) (Wu et al., 2014; Xin et al., 2015). Similarly to polysaccharides, the SA is characterized by electrostatic interactions between molecules due to carboxyl functional groups. Physical properties of SA are very susceptible to vary depending on the solution chemistry such as pH or total ionic strength change. At neutral pH, the high negative charge of SA due to deprotonated carboxylic functional groups, induces repulsive inter- and intramolecular electrostatic forces. Moreover, any change of ionic strength in a sodium alginate solution has a significant effect, especially on polymer chain extension (Ang et al., 2006; Lee et al., 2006; Lee and Elimelech, 2006; Xu et al., 2017). In the presence of divalent cations, such as Ca^{2+}, alginates form complexes with unique structure, resulting in a high density gel network (van den Brink et al., 2009; Wang and Waite, 2009; Weinman and Husson, 2016). Alginate gel formation in the presence of calcium ions has been explained by the "egg-box" model, according to which calcium ions bind preferentially to the carboxylic groups of alginate in an organized manner and form bridges between adjacent alginate molecules, leading to the egg-box-shaped gel network (Grant et al., 1973; Katsoufidou et al., 2007).

Furthermore, numerous works studied the fouling mechanisms occurring when filtering sodium alginate solution, however most of those studies focused on RO and nanofiltration (NF) membrane filtration (Ang et al., 2006; Hong and Elimelech, 1997; Lee et al., 2006; Lee and Elimelech, 2006). Ye et al. (2005b) tried to identify fouling mechanisms based on dynamic filtration experiments of sodium alginate solution using MF and UF membranes. Experimental data from both dead-end and cross-flow filtration experiments using MF membranes revealed the existence of a two-stage fouling mechanism; pore constriction in short term, while in long term fouling was found to be controlled by cake development on the membrane surface (Ye et al., 2006; Ye et al., 2005a,b). On the other hand, permeate flux decline during UF was found to be consistent with the cake filtration model throughout the duration of the experiment although adsorption or pore blocking phenomena existed during the early stages of filtration (Charfi et al., 2015; Ye et al., 2005b).

Since microfiltration for seawater desalination pretreatment is still not well studied, this work aims to better understand fouling by AOM occurring with low-pressure driven membrane such as microfiltration during seawater desalination pretreatment. For this purpose, sodium alginate microfiltration under high salinity conditions has been experimented. Moreover, identification of fouling mechanisms was made by comparing permeate flux experimental data to Hermia blocking filtration model (Hermia, 1982).

2. Method

This work aims to determine the fouling mechanism responsible for permeate flux decline when proceeding filtration of sodium alginate solution in the presence of NaCl and $CaCl_2$, using dead end filtration mode.

For fouling mechanism identification, the classical blocking laws proposed by Hermia (1982), have been used for dead end filtration at constant pressure. They consist in four fouling models shown in Table 1, describing separately four different fouling mechanisms: (i) pore constriction, (ii) complete blocking (iii) intermediate blocking and (iv) cake formation. While pore constriction model is based on the decrease of membrane pore size when foulants are adsorbed in membrane pores, the complete blocking model assumes the total sealing of membrane pores when foulants settle on membrane surface. Intermediate blocking model is based on the probability of a pore being blocked by foulant coming towards the membrane and cake formation model assumes the formation of a deposit on the membrane surface.

Where J is the permeate flux ($m^3.m^{-2}.s^{-1}$), J_0 the initial permeate flux ($m^3.m^{-2}.s^{-1}$), t the filtration time (s), C_{SA} the sodium algi-

Table 1
Hermia blocking filtration laws.

Fouling mechanism	Expression	
Pore constriction	$J = \frac{4.J_0}{(K_{pc}.J_0.t+2)^2}$ (Eq.(1))	$K_{pc} = \frac{2.C_{SA}}{\rho_f.e}$ (Eq.(2))
Complete blocking	$J = J_0 exp(-K_{cb}.t)$ (Eq.(3))	$K_{cb} = J_0.\sigma$ (Eq.(4))
Intermediate blocking	$J = \frac{J_0}{K_{ib}.J_0.t+1}$ (Eq.(5))	$K_{ib} = \sigma$ (Eq.(6))
Cake formation	$J = \frac{J_0}{(2.K_{cf}.J_0^2.t+1)^{\frac{1}{2}}}$ (Eq.(7))	$K_{cf} = \frac{\alpha_{ave}.C_{SA}.\mu}{\Delta P}$ (Eq.(8))