



Enhanced gypsum scaling by organic fouling layer on nanofiltration membrane: Characteristics and mechanisms



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ABSTRACT

To investigate how the characteristics of pregenerated organic fouling layers on nanofiltration (NF) membranes influence the subsequent gypsum scaling behavior, filtration experiments with gypsum were carried out with organic-fouled poly(piperazineamide) NF membranes. Organic fouling layer on membrane was induced by bovine serum albumin (BSA), humic acid (HA), and sodium alginate (SA), respectively. The morphology and components of the scalants, the role of Ca^{2+} adsorption on the organic fouling layer during gypsum crystallization, and the interaction forces of gypsum on the membrane surface were investigated. The results indicated that SA- and HA-fouled membranes had higher surface crystallization tendency along with more severe flux decline during gypsum scaling than BSA-fouled and virgin membranes because HA and SA macromolecules acted as nuclei for crystallization. Based on the analyses of Ca^{2+} adsorption onto organic adlayers and adhesion forces, it was found that the flux decline rate and extent in the gypsum scaling experiment was positively related to the Ca^{2+} -binding capacity of the organic matter. Although the dominant gypsum scaling mechanism was affected by coupling physicochemical effects, the controlling factors varied among foulants. Nevertheless, the carboxyl density of organic matter played an important role in determining surface crystallization on organic-fouled membrane.

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1. Introduction

With the increasing requirement for high quality water, membrane processes have attracted worldwide attention because of their potential to provide efficient and enhanced water purification as well as environmental and commercial benefit. Nanofiltration (NF) membranes are widely used in systems for seawater desalination, drinking water purification, wastewater treatment and reclamation, because they are capable of removing a broad range of organic, inorganic, and microbial contaminants in a single treatment step with their nanoscale pore size and/or surface charge (Hong and Elimelech, 1997; Van Der Bruggen et al., 2003). Removal of hardness at low operating pressure, high productivity, and low operational cost also make NF membranes one of the more favorable alternatives in the drinking water purification industry.

Although its application in the water industry is noteworthy (Mohammad et al., 2015; Van Der Bruggen et al., 2003), just like other membrane processes, NF also suffers from the enduring problem of membrane fouling caused by organic solutes, inorganic solutes, colloids, or biological solids (Mohammad et al., 2015). Membrane fouling has been a major obstacle for most applications of NF processes in the water industry, especially in cases where high concentrations of natural organic matters (NOMs) and inorganic constituents occur (Le Gouellec and Elimelech, 2002; Thorsen, 2004).

Because of the retention effect of NF membranes, the ionic concentrations near or on the NF membrane surface will increase, and might exceed the solubility limit of sparingly soluble salts. Consequently, inorganic scalants could form. This situation usually happens in membrane desalination systems with high product water recovery. The most common constituents of scale are calcium carbonate, gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), barium/strontium sulfate and silica, although other potential scalants exist (Schäfer et al., 2005).

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Among the range of sparingly soluble salts that lead to scaling in membrane processes for seawater/brackish water desalination, gypsum is one of the most ubiquitous scaling sources because of its relatively high concentration in natural waters (Dydo et al., 2003; Le Gouellec and Elimelech, 2002). However, control of gypsum is a major challenge in the development of membrane processes for desalination, since it cannot be effectively prevented by lowering feedwater pH or chemical cleaning.

As a result of scaling, membrane flux declines severely, permeate quality reduces, and the life of the membrane system is significantly shortened (Lee et al., 1999; Lee and Lee, 2000). Consequently, obtaining a full understanding of gypsum scaling mechanisms as well as how the related factors affect gypsum scaling is very important to mitigate gypsum scaling. Various parameters affecting the crystallization process have already been identified, such as temperature (Hoang et al., 2007), pH (Her et al., 2000), operating pressure (Lee and Lee, 2000), flow velocity (Lee and Lee, 2000), salt concentration (Le Gouellec and Elimelech, 2002), types of antiscalant (Rahman, 2013), coexisting sparingly soluble salts (Sheikholeslami, 2003), and other metal ions (Hamdona and Al Hadad, 2007). In addition, coexisting NOM has also been considered to interfere with the formation of gypsum precipitates (Barcelona and Atwood, 1978) and the performance of NOM on various forms of scaling on reverse osmosis (RO)/NF membranes has also been investigated (Wiesner, 2007). A previous study demonstrated that NF membrane flux decline because of gypsum scaling was much slower in the presence of 3 mg L^{-1} humic acid (HA) than in the absence of HA (Le Gouellec and Elimelech, 2002). A similar phenomenon was found in gypsum scale formation on RO/NF membranes in the presence of HA, where HA acted like an antiscalant and substantially decreased the rate of formation of gypsum scaling (Lee et al., 2009).

Organic fouling occurs widely in NF membrane processes (Hong and Elimelech, 1997; Seidel and Elimelech, 2002) because organic macromolecules are ubiquitous in all natural water sources (e.g., seawater, surface water, and groundwater). Although membrane cleaning has the ability to remove the adhesive foulants and restore the flux to various degrees (Mi and Elimelech, 2008, 2010bbib_Mi_and_Elimelech_2010b), the accumulation of organic foulants on the membrane surface over time is inevitable. It is likely that membrane organic fouling usually precedes inorganic scaling in typical NF systems (Huiting et al., 2001; Yang et al., 2008). Hence, a pregenerated organic fouling layer would probably change the physicochemical properties of the membrane surface. Consequently, the subsequent inorganic scaling behaviors will likely be affected by the specifics of the pregenerated organic fouling layer. For instance, in a forward osmosis (FO) membrane process, when gypsum coexisted with alginate, the permeate flux decreased faster than for alginate/gypsum alone; moreover, the most severe flux decline was observed when alginate was precoated on the FO membrane surface (Liu and Mi, 2012). Therefore, it is significant to investigate how the surface characteristics of NF membranes are altered by organic fouling layers and further influence gypsum scaling for the development of useful protocols for fouling mitigation. Nevertheless, to our knowledge, there has been no reported research on this aspect to date.

This study aimed to investigate the influence of pregenerated organic fouling layers on the subsequent gypsum scaling of NF membranes, and determine the characteristics of gypsum scaling behavior of diverse organic conditions as well as virgin membrane to elucidate the underlying mechanisms. Three typical organic foulants—bovine serum albumin (BSA), HA, and sodium alginate (SA)—were used to represent widespread organic foulants (proteins, humics, and polysaccharides, respectively). To distinguish the influence of the organic fouling layer in a pressure-driven filtration

process, bench-scale experiments on gypsum scaling and subsequent cleaning of different conditioned membranes were conducted and compared. Scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS) analyses of the scalants were performed to determine whether the characteristics of the membrane surface impacted the gypsum scaling morphology and chemical composition. Quartz crystal microbalance with dissipation (QCM-D) was utilized to investigate the influence of the chemical characteristics of each organic layer on the adsorption of calcium ions and quantitatively evaluate the role of Ca^{2+} adsorption on the structural characteristics of the adhesion layer. Atomic force microscopy (AFM) force measurements were conducted to determine the interaction characteristics of different organic fouling layers on gypsum scaling and to elucidate the scaling mechanisms at the nanoscale. The ultimate goal was to provide useful information about targeted removal of organic matter for feedwater pretreatment in desalination processes.

2. Materials and methods

2.1. Poly(piperazineamide) NF membrane

The membranes used in this study were poly(piperazineamide) NF membranes with polysulfone support. The detailed process of manufacture of the fabricated NF membranes is given in Section S1 of the Supporting Information. The average pure water flux of the NF membrane tested under 0.6 MPa was $48 \text{ L m}^{-2} \text{ h}^{-1}$. The divalent salt rejection was 94%, determined with a 2 g L^{-1} MgSO_4 feed solution at an applied pressure of 0.6 MPa and a cross-flow velocity of 8 cm s^{-1} . Other properties and AFM images of the prepared poly(piperazineamide) NF membrane are shown in Table S1 and Fig. S1 in Section S2.

2.2. Organic foulants and inorganic salts

Model organic foulants (BSA, HA, and SA) were purchased from Sigma–Aldrich Co. LLC (St. Louis, MO) and received in powder form. The stock solution of each organic foulant (2 g L^{-1}) was prepared as detailed in Section S3. Inorganic salts including sodium chloride (NaCl), sodium sulfate (Na_2SO_4), and calcium chloride (CaCl_2) were provided by Tianli Chemical Reagent Co. Ltd. (Tianjin, China). All inorganic reagents were analytical grade and used without further purification. Deionized (DI) water (conductivity $< 2 \mu\text{S cm}^{-1}$, pH = 7–7.5) was used throughout this study unless otherwise stated.

2.3. Protocols for the NF membrane fouling, scaling and cleaning experiments

The fouling, scaling, and cleaning experiments were conducted with a bench-scale cross-flow system that comprised a rectangular plate-and-frame membrane cell with a dimensioned rectangular channel (12 cm long, 4 cm wide, and 0.3 cm deep) inside. A schematic diagram of the setup is depicted in Fig. S2.

The protocols for the entire experiment are illustrated in Fig. 1. Each experiment included five different and consecutive stages, namely compaction, conditioning, organic fouling, gypsum scaling, and water cleaning, as follows: (i) the clean membrane was compacted and equilibrated with DI water at 0.7 MPa for at least 5 h; (ii) the membrane was conditioned under baseline conditions of around 0.6 MPa (the pressure was slightly adjusted as needed) for at least 3 h until a satisfactory steady initial baseline flux was obtained; (iii) the organic fouling experiment was continuously performed with freshly prepared organic foulant solution for 24 h to introduce an organic fouling layer on the membrane surface; (iv)

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