

Combined influence of natural organic matter (NOM) and colloidal particles on nanofiltration membrane fouling

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Abstract

The combined influence of natural organic matter (NOM) and colloidal particles on the fouling of thin-film composite nanofiltration (NF) membranes is systematically investigated. Combined fouling is compared to the individual fouling behaviors (i.e., colloid or NOM alone) with respect to fouling mechanisms and the effect of concentration factor (or recovery). Results demonstrate that (1) “cake-enhanced osmotic pressure” (CEOP) is a key fouling mechanism for individual colloidal fouling, (2) NOM–calcium complexation is the dominant factor governing individual NOM fouling, and (3) combined fouling is affected by both CEOP and NOM–calcium complexation. The extent of flux decline for combined fouling, however, is less than what inferred from additivity of the individual contributions of colloidal and NOM fouling to flux decline. This observation implies that the contributions of the fouling mechanisms appear to be relatively less significant for combined fouling compared to their separate influences on individual colloidal and NOM fouling. An increase in colloidal stability in presence of NOM and the competition between colloids and NOM for calcium are likely explanations for this behavior. It is further shown that NF membrane salt rejection increases noticeably in case of combined fouling compared to individual colloidal fouling due to the formation of an active rejecting layer by the accumulated NOM on the membrane surface. Results from combined fouling runs involving EDTA treatment confirm that both CEOP and NOM–calcium complexation take place *simultaneously*.

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1. Introduction

Nanofiltration (NF) membranes operate at a lower pressure (energy) than reverse osmosis (RO) membranes and their product water quality readily meets the existing and pending drinking water quality standards. These attributes of NF membranes, along with the availability of NF membranes with a wide range of molecular weight cutoffs and chemical properties, have accelerated their use in drinking water treatment. The continuous deterioration of membrane performance due to fouling, however, is still a major challenge for

efficient application of NF membrane technology. Of the wide range of dissolved species and suspended matter in natural waters, small colloidal particles and natural organic matter (NOM) are considered as major foulants.

Numerous studies have addressed the physical and chemical factors governing colloidal or organic fouling of pressure-driven membranes [1–9]. For salt rejecting membranes – like NF and RO – colloidal fouling is caused by the accumulation of colloidal particles on the membrane surface and formation of a cake layer. However, it has been recently demonstrated that the permeate flux decline for RO and NF membranes is not caused by the colloidal cake layer resistance, but rather due to cake-enhanced osmotic pressure (CEOP) [10–12]. These studies have demonstrated that the hindered back dif-

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fusion of salt ions within the colloid deposit layer results in elevated salt concentration and, thus, an enhanced osmotic pressure near the membrane surface. The latter leads to a substantial decrease in the net driving pressure across the membrane and, consequently, to a severe flux decline. In case of NOM fouling, the decline in permeate flux is attributed to the increase in the total hydraulic resistance contributed by the NOM fouling layer [7]. Therefore, it is the structure of the NOM fouling layer – compactness and thickness – that governs flux decline behavior during NOM fouling. The structure of the NOM fouling layer is affected by both chemical (pH, ionic strength, and divalent cations) and physical (initial permeate flux and crossflow velocity) conditions [6–9]. Among these factors, NOM–calcium complexation plays a critical role in the formation of a thick, dense, and highly resistant fouling layer [7].

To date, however, there have been no systematic studies on the combined influence of colloidal particles and NOM (i.e., when feed water contains both colloids and NOM) on NF membrane fouling. In addition, studies on the influence of feed water recovery (or concentration factor) – one of the most important factors controlling process efficiency in membrane plants – on combined fouling as well as individual colloidal and NOM fouling are also rather scarce. In membrane plants, solute (salt, colloids, and NOM) concentration in spiral-wound NF modules increases downstream the pressure vessel, with the potential to significantly influence the extent of fouling and the mechanisms involved. In addition to the adverse effects of fouling on permeate water flux, little is known about the impact of combined fouling on solute rejection and the resulting product water quality.

The objective of this paper is to investigate the factors controlling *combined* colloidal/NOM fouling of NF membranes and the relevant fouling mechanisms. Specifically, the roles of concentration factor (or recovery) and solution chemistry on the permeate flux and solute rejection behaviors are evaluated in a laboratory-scale NF unit. Emphasis is placed on demonstrating the relative contributions of cake-enhanced osmotic pressure and NOM–calcium complexation – important fouling mechanisms of individual colloidal and NOM fouling, respectively – in combined fouling.

2. Materials and methods

2.1. Reagents and chemical solutions

All chemicals were ACS grade (Fisher Scientific, Pittsburgh, PA) and were used without further purification. Chemical solutions and feed waters for the fouling experiments were prepared with deionized (DI) water (Nano Pure II, Branstead, Dubuque, IA) having a conductivity less than 0.7 $\mu\text{S}/\text{cm}$ when in equilibrium with atmospheric CO_2 . Salt stock solutions for obtaining the desired feed ionic strength, divalent (calcium) ion concentration, and pH were prepared

using NaCl , $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, and NaHCO_3 , respectively. To investigate the role of NOM–calcium complexation in fouling, a disodium EDTA ($\text{Na}_2\text{C}_{10}\text{H}_{14}\text{O}_8\text{N}_2 \cdot 2\text{H}_2\text{O}$) stock solution was prepared with DI water. Salt (NaCl) and Ca^{2+} concentrations of feed and permeate samples were measured during the course of the fouling tests using conductivity meter (Model 32, YSI Co., Inc., Yellow Spring, OH) and calcium selective electrode (Model 93-20, Orion Research Inc., Boston, MA) along with reference electrode (Model 90-01, Orion Research Inc., Boston, MA) connected to pH/mV meter (Accumet, Fisher Scientific, Pittsburgh, PA), respectively.

2.2. Preparation and characterization of the colloid and NOM foulants

Commercial colloidal silica (SiO_2) particles with an average diameter of 300 nm (MP-3040, Nissan Chemical Industries, Ltd., NY) and an initial concentration of 200 mg/l were used as the model colloidal foulant for the individual colloidal (colloids alone) and combined (colloids plus NOM) fouling experiments. The colloidal silica was supplied as a stable concentrated aqueous suspension at an alkaline pH and was used as obtained with no further treatment. Gravimetric analysis revealed the density of the particles to be 2.30 g/cm^3 . Feed colloid concentration was measured during the course of the fouling tests using turbidity meter (Model 43900, HACH Company, CA). Scanning electron microscopy (SEM) (PhotoMet, PhotoMetrics, Inc., Huntington Beach, CA) was used to examine the uniformity (or polydispersity) and shape of the model silica colloids. The silica colloids were also characterized by measuring electrophoretic mobility under solution chemistries utilized in the fouling experiments (ZetaPALS, Brookhaven Instruments Corp., New York).

The model NOM foulant was concentrated from Nakdong River (Changweon, Korea) by a pre-softening step, followed by RO concentration. The concentrated NOM was further freeze-dried to make a powder stock. Dissolved organic carbon content (DOC) of the NOM powder was determined to be 0.462 g DOC/g NOM using a TOC analyzer (Model 820, SIEVERS, Ionics Instrument Business Group, Boulder, CO). An initial concentration of 20 mg (NOM powder) per liter was used for the individual NOM (NOM alone) and combined (colloids plus NOM) fouling tests. NOM concentration of feed and permeate samples was measured during the course of the fouling tests using UV absorbance at 254 nm (Hewlett Packard 8453).

The NOM was characterized with respect to size, structure, and functionality. Molecular weight (MW) and MW distribution of NOM were determined by high-performance size-exclusion chromatography. XAD-8/4 resins were used for fractionation of hydrophobic NOM (XAD-8 adsorbable), transphilic NOM (XAD-4 adsorbable), and hydrophilic NOM (neither XAD-8 nor XAD-4 adsorbable) constituents. Carboxylic acidity (pH 3–8) and phenolic acidity (twice of

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