

Nano-colloidal fouling mechanisms in seawater reverse osmosis process evaluated by cake resistance simulator-modified fouling index nanofiltration



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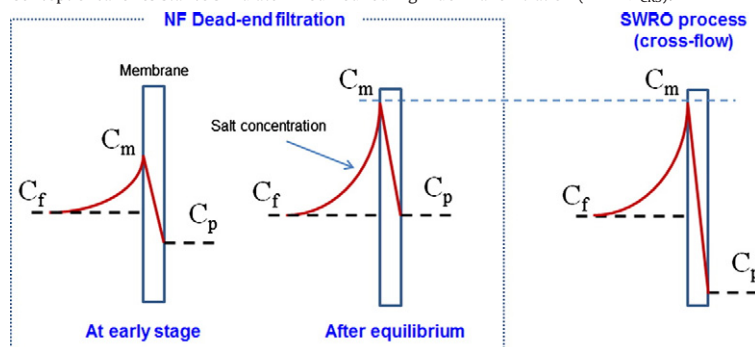
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HIGHLIGHTS

- The fouling mechanisms of nano-colloids in SWRO desalination were investigated.
- A novel method was developed to determine the specific cake resistance.
- Specific cake resistances were evaluated by cake resistance simulator (MFI-NF_{CRS}).
- Flux decline in SWRO process greatly depended on the cake-enhanced osmotic pressure.

GRAPHICAL ABSTRACT

Concept of cake resistance simulator-modified fouling index nanofiltration (MFI-NF_{CRS}).



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ABSTRACT

Fouling potentials caused by particulate matters are generally evaluated by fouling index, such as SDI and MFI, widely used in RO membrane practices. However, these fouling indices failed to predict the effect of nano-colloidal sizes on flux decline, implying that colloidal fouling is too complex to be analyzed by simple fouling index. Thus, in this study, nano-colloidal fouling mechanism in seawater reverse osmosis (SWRO) desalination was fundamentally investigated by employing new approach. Specifically, the specific cake resistance of colloidal foulants was first determined by a novel method, cake resistance simulator-modified fouling index nanofiltration (MFI-NF_{CRS}), which was conducted under operating pressure and solute environment similar to those of real SWRO desalination. Then colloidal deposition and resulting cake-enhanced osmotic pressure (CEOP) were quantitatively assessed by fitting RO experimental data to the calculations from the CEOP model. The results clearly demonstrated that the flux decline caused colloidal deposition in the SWRO process depended greatly on the CEOP. The newly developed methodology including MFI-NF_{CRS} is expected to contribute significantly to better understand nano-colloidal fouling mechanisms and to accurately predict their fouling potentials in the SWRO desalination.

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1. Introduction

There are various types of membrane fouling, such as colloidal deposition, organic adsorption, inorganic scaling and biofouling, which cause

the gradual reduction of permeate flux and the deterioration of permeate water quality. Among these foulants, colloidal deposition appears to be one of the main causes of membrane fouling in the reverse osmosis (RO) membrane process which is becoming increasingly popular for seawater desalination [1–4]. Numerous studies have addressed the physical and chemical factors affecting colloidal deposition in pressure-driven membrane systems [5–10]. Colloids are roughly

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defined as fine particles with sizes in the range of 1 nm–1 μm [11] and those are likely to cause membrane fouling as a result of their sizes [12]. In addition, a more densely packed cake layer is formed on the membrane surface in seawater reverse osmosis (SWRO) processes, due to the reduction of electrostatic double-layer repulsive forces among colloids, which consequently causes greater resistance to permeate flux [7]. Although the removal efficiency of larger particles has been improved significantly by advanced pretreatments, such as microfiltration (MF) and ultrafiltration (UF) [13], the membrane fouling caused by smaller nano-colloids still remains as a recalcitrant problem in the SWRO desalination.

The silt density index (SDI) and/or modified fouling index (MFI) are widely used to predict the fouling potential of feed water for RO processes. These indices, however, often fail to predict the extent of colloidal fouling in real SWRO processes, primarily due to the differences in membrane pore sizes and complex fouling mechanisms. Relatively new methods, namely, MFI-UF and MFI-NF, have been proposed to predict the fouling potential caused by fine colloidal particles and organic substances, employing UF or NF membranes, respectively, in MFI measurements [14–17]. Recently, a novel fouling index determined by a set of membranes with different pore sizes was developed for more accurate estimations of feed fouling potentials [18,19]. Some researchers have suggested a new approach to incorporate crossflow hydrodynamic behavior during fouling index measurements, known as the crossflow sampler modified fouling indices (CFS-MFI_{UF}) [10,20,21].

The MFI based fouling indices are derived from cake filtration theory and have a linear relationship with specific cake resistances and particle concentrations. In addition to hydraulic resistance, the cake layer formed in the RO process causes cake-enhanced osmotic pressure (CEOP), which decreases the effective pressure across RO membrane due to increasing solute concentrations at the membrane surface [22–24]. The extent of a cake layer formation is greatly affected by solution chemistry (e.g., pH and ionic strength), membrane properties, and hydrodynamic conditions (e.g., critical flux) [25]. Despite its importance, to date, a fouling index to reflect complex mechanisms of colloidal fouling in the SWRO process has not been developed yet. Recent studies have described the significant effect of the CEOP on flux decline, but their results could not be applicable to SWRO process due to lower total dissolved solids (TDS) conditions (i.e., 10 g/L NaCl or lower) at which their RO experiments were conducted [22–24].

In this study, the colloidal fouling mechanism that significantly affects the permeate flux in the SWRO process was systematically investigated through fouling index measurements and lab-scale RO experiments using nano-colloids with different sizes. Specifically, the fouling potentials of colloidal foulants were first evaluated through SDI, MFI-MF, and MFI-UF. The discrepancy between the fouling potentials measured by fouling indices and the actual flux decline of SWRO experiments was identified with respect to colloidal size, leading to a more fundamental investigation on nano-colloidal fouling mechanism in the SWRO process. The specific cake resistance of nano-colloids was first determined by a novel method, cake resistance simulator-modified fouling index nano-filtration (MFI-NF_{CRS}). More specifically, NF dead-end filtrations were conducted under the same operating pressures and solute conditions as the practical SWRO process to obtain values similar to the specific resistances of the cake layer formed in the real SWRO process. Then, colloidal fouling mechanisms were fundamentally elucidated by performing systematic theoretical and experimental analysis based cake enhanced osmotic pressure (CEOP) model and MFI-NF_{CRS}. Finally, the contributions of cake resistance and CEOP to the flux decline under seawater conditions were compared and delineated. Our observations from experiments as well as modeling provide new insights into nano-colloidal fouling mechanisms in the SWRO process and strategies for the prediction of their fouling potentials.

2. Materials and methods

2.1. Colloidal particle foulants

Commercial silica (SiO_2) particles with different sizes were used as model foulants to evaluate fouling potential with respect to particle size. The sizes of model particles were 3 μm (SILNOS-3 M, ABC Nanotech, Korea), 70–100 nm, 40–60 nm and 10–15 nm (ST-ZL, ST-XL and ST-30, Nissan Chemical Industries, Japan). The 3 μm particles were obtained in a powdered form, and the other nano-colloidal particles were supplied in a dissolved form at 40% (ST-ZL, ST-XL) or 30% (ST-30) concentrations (w/w). The feed water samples containing different concentrations of these particles were prepared by adding them to deionized (DI) water (SHUVQ-90ST, Sinhan Science Tech.) or pretreated seawater. Gravimetric analysis revealed the density of 70–100 nm, 40–60 nm and 10–15 nm particles to be 2.3, 2.1 and 2.1 g/cm^3 , respectively. The size distributions of these silica particles were measured using a particle size analyzer (Zetasizer Nano ZS, Malvern Instrument Ltd.) in DI water and 35 g/L NaCl solution.

To elucidate the effect of particle size on flux decline in the SWRO process, the sizes of nano-colloidal suspensions at the seawater-level solute conditions were measured accurately, as shown in Fig. 1. The average sizes of 10–15 nm, 40–60 nm and 70–100 nm colloidal particles increased to 28 nm, 91 nm and 164 nm, respectively, due to the aggregation of colloids in the 35 g/L NaCl solution. The average particle size of 3 μm , however, did not show any changes (i.e., 2.90 μm in DI water and 2.92 μm in the 35 g/L NaCl solution). These observations are in good agreement with the results of a previous study [10]. The high ionic strength leads to a reduction of the electrostatic double layer (EDL) repulsion among the colloids, causing particle size increase due to particle aggregation. Because the main goal of this study is to elucidate the colloidal fouling mechanism in the SWRO process, colloid sizes of 28 nm, 91 nm and 164 nm were used to evaluate the effect of particle size on nano-colloidal fouling in the following sections.

2.2. Feed water

Two types of feed water were used to investigate the mechanism of colloidal fouling in SWRO process. One was a pretreated seawater taken from the sea near Changwon, Korea. The pretreated seawater sample was collected from the feed tank of an RO pilot plant (50 m^3/day) equipped with a microfiltration membrane (i.e., pore size 0.1 μm), which served as a pretreatment process. The pretreated seawater sample was carefully delivered to the laboratory and stored in a refrigerator. The basic water quality of the sample was analyzed, and the results are listed in Table 1.

Another feed water was DI water in which the TDS was adjusted to be 35 g/L (i.e., similar to seawater TDS) by adding NaCl, and the pH was fixed at 7.0 ± 0.5 using 0.1 N NaOH or 0.1 N HCl stock solutions. Aldrich humic acid (HA) was also added to the solution to produce an organic concentration similar to seawater. The stock solution was prepared by dissolving HA in DI water, followed by stirring for 2 days. The stock solution (2000 mg/L) was filtered through a UF membrane with an MWCO of 30 kDa to remove colloidal organics, and then, the dissolved organic carbon (DOC) was determined using a TOC analyzer (TOC-V, Shimadzu Corp.). The DOC value of this feed water was adjusted to 0.5 mg/L by adding the HA stock solution.

2.3. SDI and MFI measurements

SDI measurement was performed by the standard test method (ASTM 4189-07). The SDI equipment consisted of a membrane holder, a high pressure pump controlled by pressure control system, feed water reservoir, and data acquisition system. During SDI tests, the applied pressure was kept at 2.07 ± 0.07 bar and the feed water temperature was controlled at 20 ± 1 °C through a cooling coil submerged

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