

Multiple MFI measurements for the evaluation of organic fouling in SWRO desalination



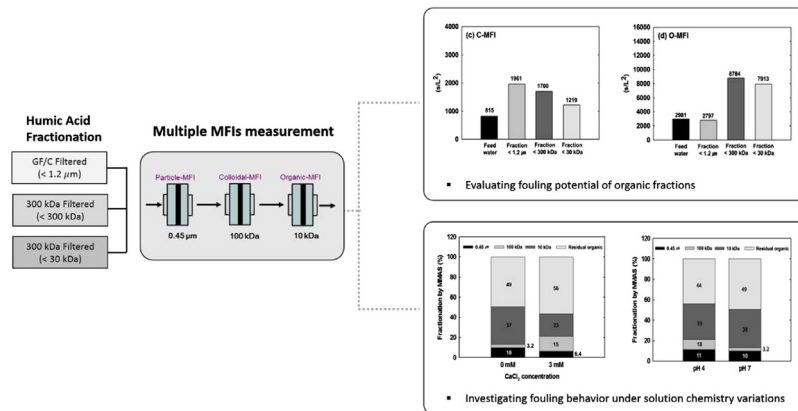
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HIGHLIGHTS

- Organic fouling potential and mechanism were evaluated accurately by multiple MFIs.
- Particle, colloid and organic MFIs were measured consecutively by MMAS.
- Humic fractions manifested by high colloid and organic MFIs caused severe fouling.
- Organic fouling elevated by low pH and high Ca^{2+} was well predicted by colloid-MFI.
- Colloid-MFI should be reduced for organic fouling control in SWRO.

GRAPHICAL ABSTRACT



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ABSTRACT

A series of MFI (modified fouling index) measurements were performed to evaluate and predict organic fouling potential and mechanism in seawater reverse osmosis (SWRO) desalination, by employing a multiple membrane array system (MMAS) in which microfiltration (MF), loose and tight ultrafiltration (UF) membranes were configured in a consecutive manner to determine the MFIs of particles, colloids, and organics. The results suggested that organic fractions manifested by colloid-MFI should be removed to control organic fouling effectively, as the organic fraction with MW ranging from 30 to 300 kDa led to the most severe fouling in RO experiments. The variations in fouling potential according to solution conditions (i.e., pH and Ca^{2+}) were also more accurately evaluated using multiple MFIs than with conventional SDI. This work demonstrated that the current fouling index, relying on a single membrane, could be improved by utilizing multiple membranes with different characteristics (e.g., pore size and hydrophobicity) to assess the fouling behavior of specific organic fractions.

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1. Introduction

There has been a growing need for alternative water resources in the regions suffering from water scarcity around the world over the last few

decades. One of the best solutions for water shortage problems is seawater desalination, especially employing reverse osmosis (RO) membrane [1,2], which has been also widely used for wastewater reclamation and water purification. However, RO membrane fouling is still the major obstacle, leading to low productivity, permeate quality deterioration, high energy consumption, and large maintenance costs [3–8]. Therefore, it has been of paramount importance to predict membrane

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fouling in advance and consequently prevent the RO membrane from severe fouling.

The organic fouling caused by dissolved organic matter has been increasingly addressed in recent studies. The amount of organic matter, of which the largest portion is humic substances (more than 70%), is usually only 3–7 mg/L or less in seawater. Nevertheless, it may act as one of the main causes of membrane fouling if not removed by proper pretreatment [9,10]. The physical and chemical factors affecting organic fouling of RO membranes have been investigated in many previous studies [11–13]. Among these factors, pH and divalent cation concentration significantly influence RO fouling behavior owing to their direct effect on major functional groups of organic matter such as carboxyl and hydroxyl groups. At a lower pH, organic fouling significantly increases due to the decreased electrostatic repulsion between organic foulants and the membrane surface as well as between organic foulants themselves. At a high concentration of divalent cation, which has a strong affinity for anionic functional groups of organic matter, severe flux declines are observed due to the neutralization of functional groups and the formation of organic-calcium complexation. Because of such complex phenomena in organic fouling, there has been difficulty in the accurate prediction of fouling potential in the RO process.

A fouling index is necessary for predicting organic fouling precisely to select proper pretreatment, and thus for mitigating the fouling problem caused by organic matter. The silt density index (SDI) and modified fouling index (MFI) are widely used to predict the fouling potential of feed water for RO processes [14]. These indices, however, often fail to estimate the organic fouling potential of practical SWRO processes owing to the differences in the pore sizes of the membranes used and the complexity of the fouling mechanisms [15–17]. Relatively new methods, namely the MFI-UF and MFI-NF, have been proposed to evaluate the fouling potential caused by fine colloidal particles and organic substances employing UF or NF membranes, respectively, in MFI measurements [18–20]. Nevertheless, MFI-UF and MFI-NF exhibit limitations in precise prediction of fouling potential in RO practices. MFIs using membranes with a smaller pore size may estimate fouling potential affected by various materials at a time, while those using membranes with a larger pore size often failed to predict membrane fouling caused by smaller foulants. Therefore, in a single MFI measurement, there is a limit in obtaining information indicating which foulants result in membrane fouling.

In our previous work, a new approach using a multiple membrane array system (MMAS), in which MF, loose and tight UF membranes were connected in series, has been developed [21]. The purpose of utilizing different membranes was to define the target foulants by reducing interruption by various foulants during measurement. In this configuration, the larger foulants in the feed water were filtered by the first (MF) membrane, followed by additional filtration of colloids with the second (loose UF) membrane. Finally, organic matter was filtered and measured at the third (tight UF) membrane. By determining the modified fouling index (MFI) during each separation, it was possible to calculate the Particle-MFI (P-MFI), Colloid-MFI (C-MFI), and Organic-MFI (O-MFI). Since this approach allowed for both the separation of foulants and the evaluation of the fouling potential, the major cause of the fouling with respect to the type of foulant(s) could be identified by comparing three MFI values.

The objective of this study was to improve the current single MFI method by employing the concept of multiple MFIs designed to assess the fouling potentials of particles, colloids, and organics. Specifically, organic fouling behaviors under various solution conditions were systematically investigated to demonstrate how to evaluate and predict organic fouling potential in the SWRO process by a series of MFI measurements using the MMAS. It was shown that the multiple MFI measurements more precisely reflect the changes in organic fouling potential as a result of different pretreatment options, characteristics of foulants, and solution chemistry, while the

conventional SDI and MFI measurements were shown to be insensitive in detecting these variations. Consequently, our observations provide new insights into the organic fouling mechanisms in the SWRO process as well as strategies for the reduction of fouling potential.

2. Material and methods

2.1. Organic foulants

Aldrich humic acid (HA) was used as a model foulant to simulate the organic fouling of the actual RO process. The stock solution was prepared by dissolving HA in DI water, followed by stirring for 2 days at pH 7.0 (± 0.5) adjusted using 0.1 N NaOH solution. The stock solution adjusted to 2000 mg/L was separated using three membranes with different pore sizes to investigate the effect of the molecular weight of organic foulants on RO flux decline and fouling potential as measured by MMAS. As shown in Fig. 1, the membrane types used for this purpose were a GF/C filter with a pore size of 1.2 μm (GF/C, Whatman, England), a UF with an MWCO of 300 kDa (PBMK, Millipore, U.S.), and a UF with an MWCO of 30 kDa (PLTK, Millipore, U.S.). The dissolved organic carbon (DOC) concentrations of the separated HA fractions were determined using a TOC (Total Organic Carbon) analyzer (TOC-V, Shimadzu Corp., Japan). The DOC values of feed water samples used for RO fouling runs and MMAS measurements were adjusted to target concentrations by adding each stock solution of HA fraction.

In order to investigate the effect of the addition of Ca^{2+} and the change in pH on molecular size distribution of humic acids, a fractionation method was employed [9]. The membranes used for the fractionation were an MF membrane with a pore size of 0.45 μm (HAWP, Millipore, U.S.), a UF membrane with an MWCO of 100 kDa (PLHK, Millipore, U.S.), and a UF membrane with an MWCO of 10 kDa (PLGC, Millipore, U.S.).

2.2. Feed water

Two types of feed water were used. One was a pretreated seawater sample taken from an RO pilot plant located by the sea near Changwon, Korea. The pretreated seawater sample with TOC of 1.24 mg/L was collected after passing through microfiltration membranes with a pore size of approximately 0.1 μm . It was carefully delivered to the laboratory and refrigerated. The other feed water was NaCl-spiked DI water in which the TDS was adjusted to 35 g/L to replicate seawater-level TDS. The feed water pH as well as the calcium concentration were varied (i.e., pH 4 and 7; $\text{Ca}^{2+} = 0, 0.05, \text{ and } 3.0 \text{ mM}$). The TDS, pH, and calcium concentration of each feed water were adjusted using NaCl, 0.1 N NaOH (or 0.1 N HCl), and CaCl_2 stock solutions.

2.3. Measurements of SDI and multiple MFIs

The multiple membrane array system (MMAS) test unit is presented schematically in Fig. 2. Three identical membrane cells with different membranes were connected in series. In this configuration, the fouling potential of larger foulants in feed water was evaluated at the first MF membrane with a pore size of 0.45 μm (HAWP, Millipore, U.S.) followed by additional evaluation of nano-colloids at the UF membrane with an MWCO of 100 kDa (PLHK, Millipore, U.S.). Finally, organic materials were evaluated in the UF membrane with an MWCO of 10 kDa (PLGC, Millipore, U.S.). By determining the modified fouling index (MFI) during each evaluation, Particle-MFI (P-MFI), Colloid-MFI (C-MFI), and Organic-MFI (O-MFI) were calculated and the corresponding target foulants were separated simultaneously. The volume of permeate flow during the MMAS measurements was recorded every second for 20 min in the MF membrane and for 1 h in the UF membranes. During the Particle-MFI tests, the applied pressure was kept at $2.07 \pm$

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