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# Fouling potential evaluation by cake fouling index: Theoretical development, measurements, and its implications for fouling mechanisms

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ABSTRACT

Current fouling indices typically employed in RO practices, such as silt density index (SDI) and modified fouling index (MFI), have suffered greatly from their inability to predict actual fouling potential primarily due to the erroneous interpretation of fouling mechanisms. Our findings clearly demonstrated that the effect of pore blocking should be excluded during fouling index measurements to simulate real RO applications. Thus, new concept of cake fouling index (CFI) was developed in order to accurately evaluate true fouling cake layer resistance. Specifically, the CFI was determined through consecutive filtration tests by subtracting the flux decline of the secondary filtration from that of the first one to eliminate the effect of pore blocking. The results proved that CFI better predicts the degree of fouling rate in RO experiments than MFI. It was also revealed that it could be utilized as a useful tool for identifying and evaluating the fouling mechanisms. Through a comparison of MFI and CFI, it was shown that pore blocking was enhanced as much as cake formation when pH decreased, while divalent cations ($Ca^{2+}$) increased only cake formation on the membrane surface. This newly developed index refining existing MFI method is expected to provide more precise information about RO membrane fouling, especially for the design of effective pretreatment processes.



## 1. Introduction

As a leading technology, the reverse osmosis (RO) membrane is now widely adopted for many desalting applications including seawater desalination. However, fouling is a major obstacle for its successful operation [1–4]. Several strategies to control fouling have been traditionally utilized such as improvement of membrane properties optimization of operating parameters, design of proper pretreatment process, and development of effective membrane cleaning techniques [5–7]. The implementation of these methods, however, require the precise evaluation of fouling potential and mechanism, which emphasizes the significance of fouling index development for the various RO applications. Nevertheless, current fouling indices widely employed in RO practices often fail to accurately evaluate fouling potential because of their inability to simulate actual fouling mechanisms.

In the past few decades, the fouling index measurement techniques were used as a useful membrane fouling prediction tool by employing MF or loose or tight UF membranes in RO process. The silt density index (SDI) was the most common and widely accepted tool for prediction of membrane fouling [8,9]. However, the SDI protocol is based on a 15-min pore-plugging dead-end filtration, so it is impossible to simulate real RO operation and to offer further information on foulants and/or fouling mechanisms. Furthermore, the non-linearity of foulant concentration was asserted as a limitation of SDI [10–13]. Therefore, fouling indices that reasonably trace the reflection of fouling mechanisms have been developed to replace SDI.

The modified fouling index (MFI) was suggested to measure the rate of cake formation on the membrane surface, which allows for better prediction of RO fouling phenomena. The basic concept of MFI uses the assumption that fouling mechanisms occur in the order of pore blocking, cake filtration, and then cake compression, determining the fouling index through a classical filtration model [14]. In order to simulate nonporous RO performance, the MFI was developed with the replacement of membranes with smaller pore sizes, the MFI-UF and MFI-NF of which having recently been actively studied [15–17]. Finally, Yu et al. [18,19] addressed the threshold of single MFI to verify fouling potential and developed a new concept, the multiple membrane array system (MMAS). MMAS configures series of membranes with different pore sizes (i.e., MF, UF, and NF) for assortment of fouling potential according to the characteristics of foulants. In fact, this subdivided MFI has demonstrated well the enhancement of water quality through pretreatment whereas the SDI could not reflect the change that was revealed through lab-scale RO experiments.

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Even though the MFI is regarded as an effective tool for measuring fouling potential, there is still an argument that this should be determined through a pore blocking model not cake formation depending on the pore size of membranes used in these measurements [9,20,21]. However, as fouling indices aim for improvement of the nonporous RO or NF processes, which are heavily affected by cake formation on the membrane surface, the MFI has to focus on cake layer resistance as a current cake filtration model. We could achieve it through the contrivance excluding pore blocking effect.

In this study, a novel method, the cake fouling index (CFI), was established, further developing the MFI concept, for a more precise evaluation of cake layer resistance. By performing consecutive filtration tests, we devised the observation of pore blocking, and thus measurements of true cake formation effect from the difference in flux declinations between two tests. In addition, by analyzing the differences between the CFI and MFI, fouling mechanisms were revealed more fundamentally. In particular, the behavior of the humic effect according to the chemical condition (i.e., pH, divalent cation, and ionic strength) was studied. Our observations provide new insights into the refinement of MFI measurements in response to fouling mechanisms.

## 2. Materials and methods

### 2.1. Model foulants

In this study, commercial aldrich humic acid (AHA) obtained from the International Humic Substances Society (IHSS) was chosen as a model fouling agent. The stock solution of AHA (0.2 g/L) was prepared by dissolving the powdered form of AHA with deionized water and filtering it through a GF/C (Whatman Ltd., Kent, UK) membrane. Carrier solutions were adjusted to 600 mM (Junsei Chemical Co., Tokyo, Japan) to satisfy seawater condition. The concentration of the stock solution was confirmed by measuring the total organic carbon (TOC) value using a TOC analyzer (Shimadzu Corporation, Kyoto, Japan). The chemical composition (i.e., pH, ionic strength and hardness concentration) of feed waters were adjusted with HCl (0.1 M), NaOH (0.1 M), NaCl (10 mM and 600 mM), and $CaCl_2$ (1.5 mM and 3 mM), as needed. The temperature of samples during the fouling indices experiments was maintained at 20 °C

### 2.2. RO fouling experiments

Lab-scale RO fouling experiments were performed using commercial RO membranes (Hydranautics SWC-5 Oceanside, CA) in the crossflow filtration at an applied pressure of 50 bar. This RO membrane was stored in deionized water at 4 °C with the water replaced regularly prior to experiments. Specific information on RO membrane characteristics in terms of surface roughness, zeta potential, and contact angle are 127.33 nm, −18.04 mV, and 72.30°, respectively. Thus, this SWC-5 showed that the membrane surface was relatively rough, less charged, and hydrophobic [22].

Typical lab-scale of the RO test unit was employed in the fouling experiments. This crossflow flat filtration system was configured using the cross flow of the membrane cell, high pressure pump, stirrer, feed reservoir, chiller, and data acquisition system. The effective membrane surface area was $1.5 \times 10^{-2}$ m$^2$ and the channel length, width and height were 13 cm, 4 cm, and 0.25 cm, respectively. Operating parameters such as filtrate flux, crossflow velocity and pressure were carefully controlled by manipulation of the back-pressure regulator and bypass valve.

### 2.3. Cake fouling index measurements

The multiple membrane array system (MMAS) was employed to perform filtration experiments for fouling index measurements [19]. The MMAS was configured using various membranes (i.e., different pore sizes and materials), allocated in series to separate target foulants as well as evaluate the fouling potential of feedwater simultaneously. We selected a mixed cellulose-esters (CE) membrane (HAWP, Millipore Corp., Bedford, MA) with 0.45 μm pore size and 47 mm diameter ($13.8 \times 10^{-4}$ m$^2$) designed for dead-end filtration. The MMAS unit consisted of membrane holders, a high-pressure pump, stirrer, feed reservoir, temperature control system, and data acquisition systems. The feed water temperature was controlled by circulation of chilled water through a stainless-steel coil immersed in the feed water reservoir at approximately 20 °C. The operation of MMAS ran at constant pressure and dead-end mode.

The conceptual design for determining the cake fouling index (CFI) is schematically presented in Fig. 1. As the figure shows, permeated water was entered into the filtration process repeatedly in order to understand the effect of foulants that passed through during the first filtration test (Fig. 1 (a)). Since foulants larger than the pore size of the membrane were filtered through the first filtration process, the flux decline in the second filtering could be derived by a pore blocking effect. After that, the only pore blocking mechanism could be regarded as a factor of flux declination. The feed water was placed into 20 L reservoir and stirred continuously to prevent foulants from settling. The first permeated water from MFI measurement (Filtration 1) was gathered to 20 L reservoir and a measurement was performed in the same manner (Filtration 2) (Fig. 1 (b)). The permeated volume was recorded every 1 s for 15 min the MF during CFI measurements. The equipment was cleaned using HCl and NaOH to remove the residue of organic foulants prior to new experiments.

## 3. Results and discussion

### 3.1. Theoretical model development: cake fouling index (CFI)

To predict the fouling potential in RO applications, the fouling index should satisfy two critical points: short time measurement and high sensitivity responding to varying feed water quality. The flux decline induced by fouling needs to be evaluated quickly and accurately in the real-scale RO operation. However, fouling index employing RO membrane itself is not sensitive enough to detect the effect of varying feed water quality because of large membrane resistance compared fouling resistance, particularly in the initial stage of fouling. Thus, the porous membrane such as MF and/or UF membranes has been adopted for fouling index measurements in real RO practice. For example, the MF of 0.45 $\mu m$ (ASTM: $0.86 \times 10^{10}$ m$^{-1}$ $< R_m <$ $1.72 \times 10^{10}$ m$^{-1}$) is used in the SDI measurement which is the most widely used fouling index in the current RO industry.

As one of the fouling indices, the modified fouling index (MFI) was suggested for evaluating the rate of cake resistance based on Darcy's law and Hermia's classical filtration model. However, from the point of practical evaluation, typical MFI has shortcomings in that it may provide ambiguous values which possibly overestimate real fouling potential. This is because, different from what has been assumed, fouling mechanisms (i.e., pore blocking, cake filtration, and cake composition) occur simultaneously, eventually resulting in MFI including pore blocking in addition to cake formation. Therefore, the current study suggests cake fouling index (CFI) which is developed through the concept of MFI with consecutive filtrations in order to exclude the effect of pore blocking.