



Evaluation on membrane fouling by hydrophobic and hydrophilic substances through permeation coefficient and concentration polarization factor in SWRO processes

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Received 11 April 2018; Accepted 23 June 2018

ABSTRACT

It is difficult to evaluate fouling of high-pressure membranes such as seawater reverse osmosis (SWRO) membranes, although the fouling of low-pressure membranes such as microfiltration (MF) and ultrafiltration (UF) membranes can be evaluated by monitoring the decrease in flux or the increase in trans-membrane pressure (TMP). In particular, it is more difficult to evaluate membrane fouling of SWRO membrane in actually plant because SWRO membranes are connected in series of six to eight elements in one vessel and fouling and concentration polarization occur at the same time. This study aimed to distinguish concentration polarization from membrane fouling by calculating the permeation coefficient and concentration polarization factor with the progression of membrane fouling. As fouling was progressed by organic matters, the permeation coefficient and concentration polarization factor decreased. Fouling layers by organic matters reduce the concentration polarization factor because fouling layers reduce the permeation coefficient by increasing the filtration resistance and cause cake reduced concentration polarization (CRCP) by interrupting the convection of salt. Furthermore, membranes with forward osmotic backwashing (FOB) are effective in controlling membrane fouling because FOB alleviate the decreasing rate of the permeation coefficient and concentration polarization factor compared to membranes without FOB. Therefore, it was possible to evaluate the degree of membrane fouling in SWRO membranes by analyzing changes in the permeation coefficient and concentration polarization factor with the progression of membrane fouling and to evaluate cleaning efficiency by comparing them before and after FOB. This made it possible to use permeation coefficient and concentration polarization factor as an indicator to determine the timing of cleaning-in-place (CIP) or physical cleaning such as FOB.

Keywords: Permeation coefficient; Concentration polarization factor; Fouling; Reverse osmosis; Forward osmotic backwashing

1. Introduction

Membrane fouling also occurs in reverse osmosis membranes as low-pressure membranes such as microfiltration (MF) and ultrafiltration (UF) membranes and has a large negative impact on the overall system [1–7].

Membrane fouling in seawater reverse osmosis (SWRO) occurs on the membrane surface to reduce the membrane performance, and biofouling and silica scaling cause irreversible damage to membranes [8–11]. In particular, although it is possible to evaluate membrane fouling in low-pressure membranes due to a decrease in flux or an increase in trans-membrane pressure (TMP), it is difficult to monitor and evaluate membrane fouling in high-pressure membranes such as SWRO membranes [12,13]. This

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is because fouling and concentration polarization occur at the same time in recently developed and commercialized SWRO membranes, which has low resistance and high permeability, compared to initially developed and commercialized SWRO membranes [12,14–16]. In addition, it is inefficient to measure permeate flux to evaluate the cleaning efficiency because operating period may be shortened after cleaning and foulants are not completely removed even if flux is completely recovered by cleaning. With regard to actually used SWRO membranes, six to eight elements are connected in series in one vessel and osmotic pressure increases more because inlet feed water is concentrated through flow channels on the feed water side. As a result, it becomes more difficult to distinguish the effects of fouling from those of osmotic pressure [17–19].

Chong et al. conducted a study to distinguish concentration polarization from membrane fouling by spiking NaCl [20]. They aimed to monitor fouling by fouling membranes using colloidal silica and alginic acid, injecting NaCl, and deriving concentration polarization (CP) coefficient and the filtration resistance by fouling (R_f) through a cake filtration resistance equation (Eq. (1)) considering osmotic pressure.

$$J_v = \frac{TMP_s - CP \times \Delta\pi_{bs}}{\mu(R_m + R_f)} \quad (1)$$

However, fouling monitoring methods by measuring CP coefficient has the following two major disadvantages: 1) NaCl should be injected at every measurement and 2) this method is incorrect because membrane performance (R_m) and viscosity coefficient (μ) change when adding NaCl.

Song et al. reported that SWRO performance was controlled by thermodynamic equilibrium regions because osmotic pressure increased along flow channels in full-scale RO processes due to long channels [21]. In addition, they reported that SWRO performance was controlled by mass transfer at low pressure and by thermodynamic equilibrium regions at high pressure. In other words, flux was constant even though fouling occurred in the early stage at high pressure and membrane resistance increased, and flux decreased when membrane filtration resistance exceeded a constant value.

Tay and Song aimed to evaluate membrane fouling by introducing the concept of filtration coefficient (F) and fouling index (I_f) in full-scale RO processes [14]. I_f changes if F (F_f) is measured when the initial F (F_i) value and membrane fouling progress. I_f ranges from 0 to 1. Membrane fouling will occur less frequently as I_f approaches 0, and it will occur more frequently as I_f approaches 1. Although the filtration coefficient has the advantage of explaining the whole system with one value and deriving extent of RO membrane fouling by simple experiments, their study has the disadvantage of having difficulty in distinguishing fouling from concentration polarization.

Thus, although membrane fouling affects the decrease in RO membrane flux, increase in TMP, and decrease in salt rejection rate, there is a need to distinguish concentration polarization from membrane fouling and to monitor membrane fouling. Therefore, this study calculated the permeation coefficient (L_p) and concentration polarization factor (f_{cp}) with the progression of membrane fouling after forcibly injecting HA of hydrophobic organic matter and SA of hydrophilic organic matter at high concentrations to accelerate membrane fouling [22,23].

Table 1
SWRO membrane specifications

Model	RE2521-SR
Effective membrane area, m ²	1.1
Permeate flow rate, m ³ /d	0.85
Stabilized salt rejection, %	99.6
Element configuration	Spiral-wound
Surface charge	Negative
Membrane material	Polyamide

The stated performance is initial data taken after 30 min of operation based on the following conditions; 32,000 mg/L NaCl solution at 55 bar applied pressure, 8% recovery, 25°C and pH 6.5–7.0.

This study aimed to distinguish concentration polarization from membrane fouling by calculating the permeation coefficient, concentration polarization factor and filtration resistance by membrane fouling (R_f) with the progression of membrane fouling.

A 2.5-inch SWRO spiral wounded membrane produced by Company W (Model: RE2521-SR) among commercialized RO membranes was used in the experiments. The detailed membrane specifications are shown in Table 1.

2. Materials and methods

A 2.5-inch SWRO spiral wounded membrane produced by Company W (Model: RE2521-SR) among commercialized RO membranes was used in the experiments. The detailed membrane specifications are shown in Table 1.

The used RO membrane experimental apparatus consisted of lab-scale cross-flow RO membrane test unit. The high-pressure pump, agitator, chiller, digital pressure gauge, and flow meter were integrated to make possible automatic and continuous operation. The permeate flux of permeate water was measured using balance, and the data were automatically stored (Fig. 1). In addition, a non-corrosive high-pressure pump (SUS-316) was used to produce permeate water even in seawater, and constant-pressure operation was realized even in long-time operation by installing a relief valve and building by-pass lines immediately before flowing into RO membranes. The concentrated water line was set to inflow into the feed water tank again, and the valve was set so that the permeate water could be circulated to the feed water tank. A chiller (constant-temperature water tank) and agitator were installed in the feed water tank so that the conditions of the feed water could be constantly maintained. The circulation flow rate was measured by a flow meter of the brine line at operating pressure of 10–50 bar. RO vessels and all pipes were made of SUS-316 to prevent corrosion, and the cross-flow velocity was stably maintained at 1 L/min.

The degree of membrane fouling was evaluated by periodically calculating the permeation coefficient and concentration polarization factor with the progression of membrane fouling, and the cleaning efficiency was evaluated by calculating the permeation coefficient and concentration polarization factor before and after forward osmotic backwashing (FOB). The permeation coefficient and concentration polarization factor were derived by changing the

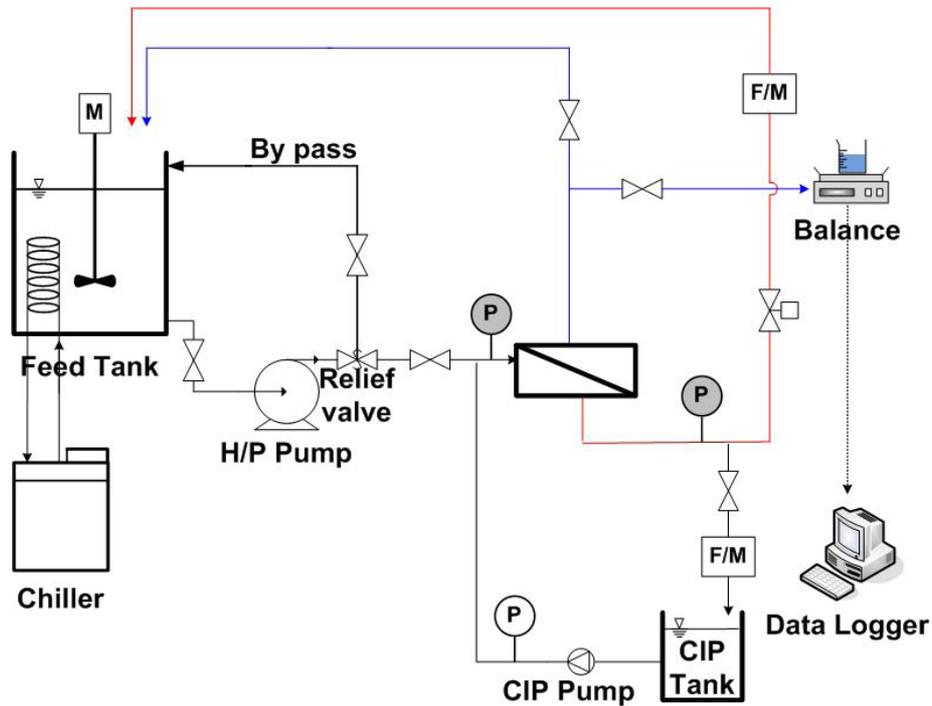


Fig. 1. Schematic diagram of experimental device for lab-scale continuous RO membrane.

thermodynamic model equation based on three assumptions as shown in Eq. (2) [22,23].

$$J = L_p(\Delta P - f_{cp}\pi_b) \quad (2)$$

where J , L_p , ΔP , f_{cp} , and π_b refer to permeate flux, permeation coefficient, trans-membrane pressure, concentration polarization factor, and osmotic pressure of inlet water, respectively. The operation was conducted under fixed conditions at 30 g/L TDS and an operating pressure of 45 bar. After adjusting TDS to 30 g/L by injecting NaCl into deionized water, TDS was increased by 0.2 g/L to measure each flux. The permeation coefficient and concentration polarization factor were calculated by measuring flux until TDS reached 30.8 g/L [23]. Organic foulants were injected in the early stage, only NaCl was injected for 48 h until it was stabilized, and then the permeation coefficient and concentration polarization factor were calculated to apply the average value as initial permeation coefficient and concentration polarization factor. Subsequently, membrane fouling was evaluated by periodically measuring the permeation coefficient and concentration polarization factor and analyzing changes in these values compared to the initial values.

Moreover, this study aimed to evaluate the degree of membrane fouling by calculating the filtration resistance by membrane fouling (R_f) from Eq. (3) after calculating the permeation coefficient and concentration polarization factor with the progression of membrane fouling.

$$J = L_p(\Delta P - f_{cp}\pi_b) = \frac{(\Delta P - f_{cp}\pi_b)}{\mu(R_m + R_f)} \quad (3)$$

where μ , R_m , and R_f refer to the viscosity coefficient of feed water, filtration resistance by membrane itself, and filtration resistance by membrane fouling, respectively.

The operation was conducted under fixed conditions at 30 g/L TDS and an operating pressure of 45 bar. Humic acid (HA) and sodium alginate (SA) represent extracellular polymeric substances (EPS) and natural organic matter (NOM), respectively. HA is a typical hydrophobic substance, and SA is a typical hydrophilic substance [24]. The used HA was filtered through a 0.45 μm filter after dissolving in deionized water, and the used SA was filtered through a 0.45 μm filter after completely stirring and dissolving it using a stirrer for more than 24 h. Both of the applied HA and SA were supplied by Sigma-Aldrich. The organic matter concentration in the feed water tank was stably maintained by removing and injecting 5 L of feed water periodically.

The experimental apparatus consisted of two series with and without FOB. FOB removes foulants attached to the membrane surface by removing operating pressure and running permeate water to feed water side using osmotic pressure. FOB has been actively studied because it is effective in moderate fouling and is environmentally friendly [1,4,5,25–27]. In this study, FOB, a physical cleaning method using osmotic pressure, was performed by reducing the operating pressure to 0 [4], and the operating pressure was rapidly reduced to 0 to minimize the effect of the conversion process in converting from filtration to FOB.

3. Results and discussion

3.1. Membrane fouling by hydrophilic organic matter

Under fixed conditions at 30 g/L of TDS concentration in feed water and an operating pressure of 45 bar, 50 mg/L SA of hydrophilic organic foulant was injected, and then changes in permeate flux, permeation coefficient (L_p), con-

centration polarization factor (f_{cp}), and filtration resistance by fouling (R_f) were observed with the passage of time.

Although the same membranes were applied to Series 1 and 2, each series had different filtration resistance (Series 1: $1.996E+14 \text{ m}^{-1}$, Series 2: $2.510E+14 \text{ m}^{-1}$) and permeate flux was relatively high in Series 1. Permeate flux in Series 1 gradually decreased by 4.14% compared to the initial flux, and permeate flux in Series 2 decreased by 1.73% compared to the initial flux (before the final backwash) by applying FOB once a day and showed a relatively low decrease rate (figure omitted).

As shown in Fig. 2, the permeation coefficient was high in membranes in Series 1 with low membrane filtration resistance, but it was relatively stable in Series 2 with periodical FOB. Although permeation coefficient in Series 1 slightly increased in the early stage, it decreased continuously by 15.20% after 21 h compared to the initial value. The permeation coefficient in Series 2 showed a reduction rate of 13.91% compared to the initial, and reduction rate of the permeation coefficient with FOB was lower than that of the case in Series 1 without FOB. The permeation coefficient decreased with increasing filtration resistance by fouling. This was because fouling was effectively controlled by FOB and then the decreasing rate of the permeation coefficient was lowered.

Fig. 3 shows the changes in the concentration polarization factor, it showed a similar tendency to the permeation coefficient. It also decreases with the progression of membrane fouling. This is similar to a phenomenon that concentration polarization factor decreases with increasing TDS concentration of feed water at constant pressure [23]. In other words, permeate flux decreases with the progression of membrane fouling on the membrane surface (constant-pressure method), and then concentration polarization is reduced. As a result, concentration polarization coefficient decreases. According to Kim et al., fouling layers by HA and SA interrupt the convection of salt and causes cake reduced concentration polarization (CRCP). The results of this study agree with their results [28]. On the other hand, when applying FOB, a membrane fouling layer is desorbed from the membrane, and then permeate flux increases. Accordingly, concentration polarization factor

increases because concentration polarization is enhanced again. Therefore, the decreasing rate of concentration polarization factor is lowered in 2 series applying FOB.

Fig. 4 shows the filtration resistance by fouling using SA as foulant. The viscosity coefficient for salt (μ_{cp}) was calculated by the ratio of the total filtration resistance (R_t) and membrane resistance (R_m) in the early stage (without membrane fouling). It was assumed that this viscosity coefficient was constant. The increasing filtration resistance with the progression of membrane fouling is the filtration resistance by fouling (R_f) because R_m is constant. The filtration resistance by fouling increased in membranes without FOB in Series 1, and the increasing tendency decreased in membranes with FOB in Series 2.

3.2. Membrane fouling by hydrophobic organic matter

Under the above conditions, 50 mg/L humic acid of hydrophobic organic foulant was injected, and then

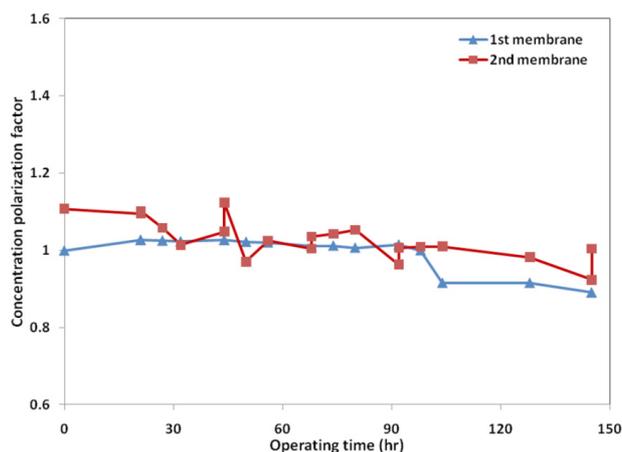


Fig. 3. Changes in concentration polarization coefficient with the passage of time (Conditions: TDS 30 g/L, operating pressure of 45 bar, injection of SA 50 mg/L).

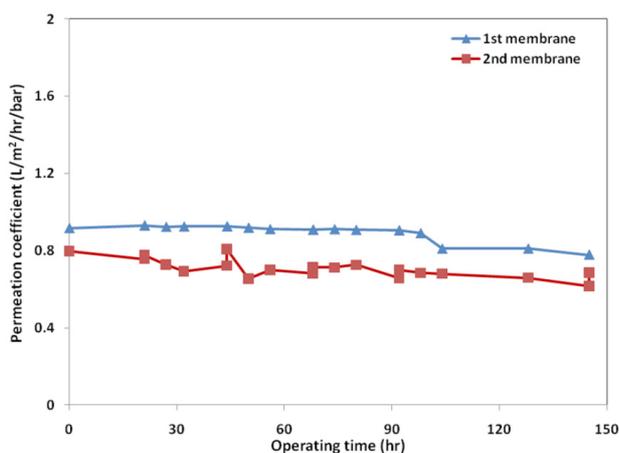


Fig. 2. Changes in permeability coefficient with the passage of time (Conditions: TDS 30 g/L, operating pressure of 45 bar, injection of SA 50 mg/L).

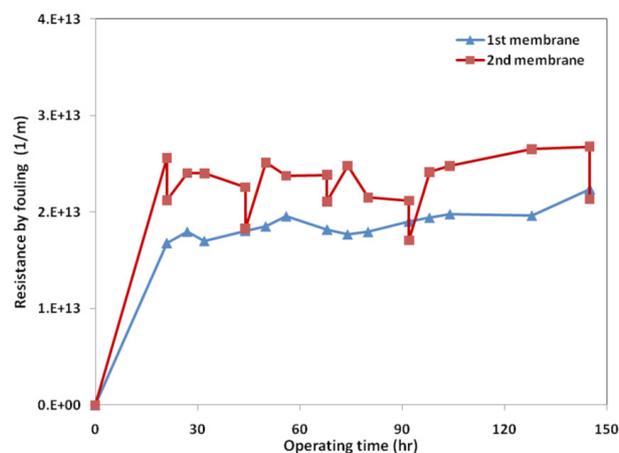


Fig. 4. Changes in filtration resistance by fouling with the passage of time (Conditions: TDS 30 g/L, operating pressure of 45 bar, injection of SA 50 mg/L).

changes in permeate flux, permeation coefficient (L_p), concentration polarization factor (f_{cp}), and filtration resistance by fouling (R_f) were observed with the passage of time. FOB was applied only to Series 1.

The permeate flux was high in membranes of Series 1 with low R_m . Although the permeate flux decreased by 0.89% compared to the initial value by repeatedly increasing and decreasing due to the application of FOB in Series 1, it decreased by 2.55% compared to the initial value by continuously decreasing in Series 2 (figure omitted). As shown in Fig. 5, the permeation coefficient was higher than the initial value in membranes in Series 1 with FOB, but it continuously decreased by 3.7% compared to the initial value in membranes in Series 2. The concentration polarization factor showed a similar tendency to the permeability coefficient. According to Fig. 6, the concentration polarization factor increased by 9.07% compared to the initial value in Series 1 with FOB, but it was maintained without major changes in Series 2. Fig. 7 is a graph showing the filtration resistance

by fouling. Likewise, membrane fouling was effectively removed in Series 1 with FOB and lower than Series 2.

3.3. Membrane fouling by hydrophilic and hydrophobic organic matter

Under the above conditions, 50 mg/L SA and HA were injected, and then changes in permeate flux, permeability coefficient (L_p), concentration polarization factor (f_{cp}), and filtration resistance by fouling (R_f) were observed with the passage of time. FOB was applied only to Series 1.

Although the permeate flux decreased by 1.97% compared to the initial value by repeatedly increasing and decreasing due to the application of FOB in Series 1, it decreased by 4.80% compared to the initial value by continuously decreasing in Series 2 (figure omitted). As shown in Fig. 8, the permeability coefficient increased as high as the initial value in membranes in Series 1 after FOB, but it continuously decreased by 20.95% compared to the initial value

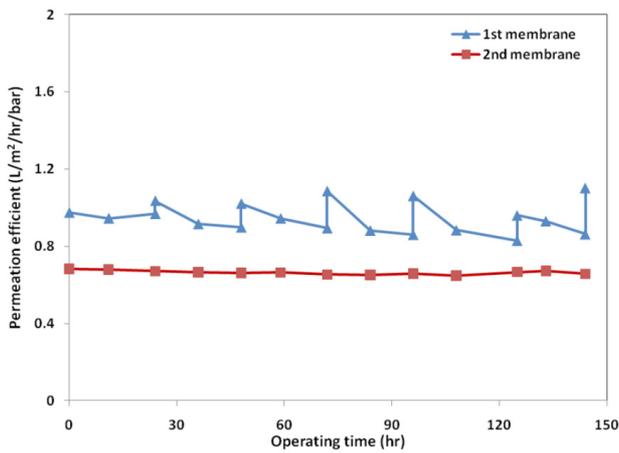


Fig. 5. Changes in permeability coefficient with the passage of time (Conditions: TDS 30 g/L, operating pressure of 45 bar, injection of HA 50 mg/L).

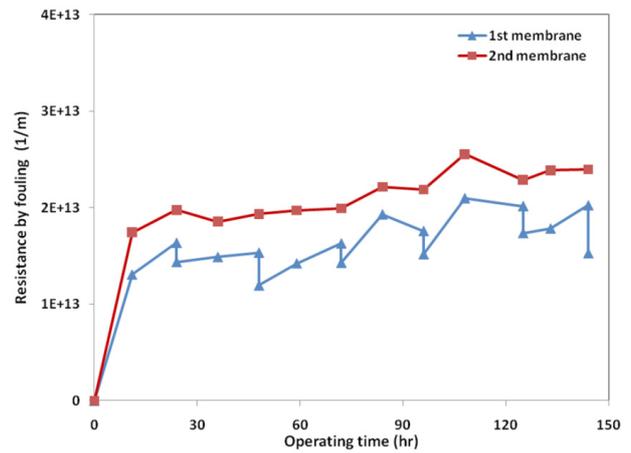


Fig. 7. Changes in filtration resistance by fouling with the passage of time (Conditions: TDS 30 g/L, operating pressure of 45 bar, injection of HA 50 mg/L).

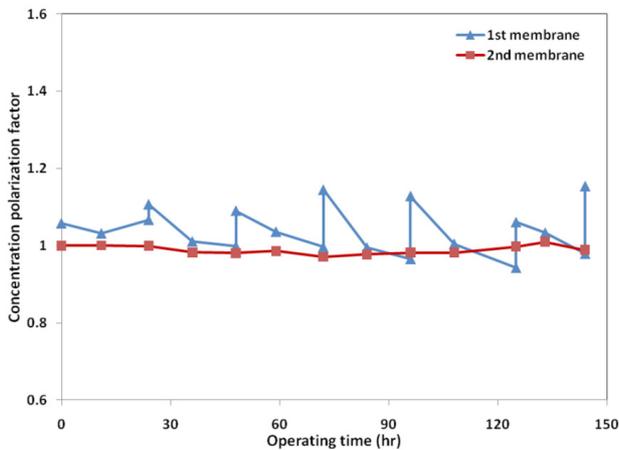


Fig. 6. Changes in concentration polarization coefficient with the passage of time (Conditions: TDS 30 g/L, operating pressure of 45 bar, injection of HA 50 mg/L).

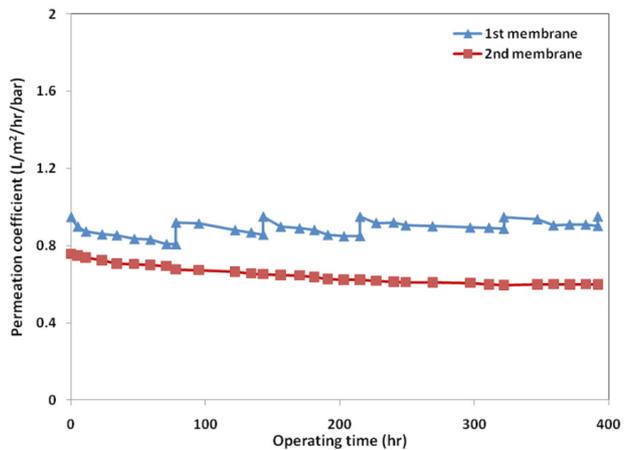


Fig. 8. Changes in permeability coefficient with the passage of time (Conditions: TDS 30 g/L, operating pressure of 45 bar, injection of HA 50 mg/L+SA 50 mg/L).

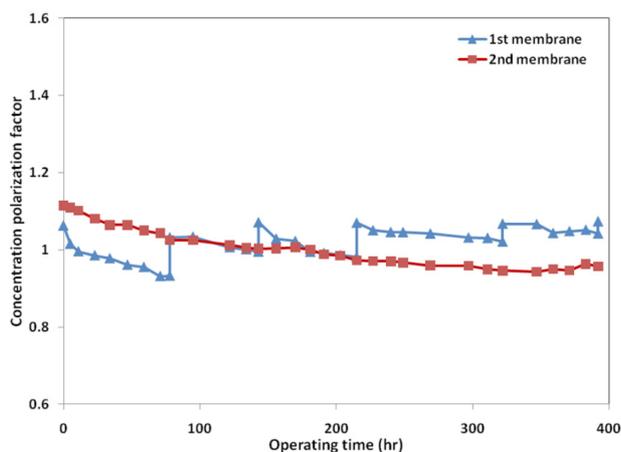


Fig. 9. Changes in concentration polarization coefficient with the passage of time (Conditions: TDS 30 g/L, operating pressure of 45 bar, injection of HA 50 mg/L+SA 50 mg/L).

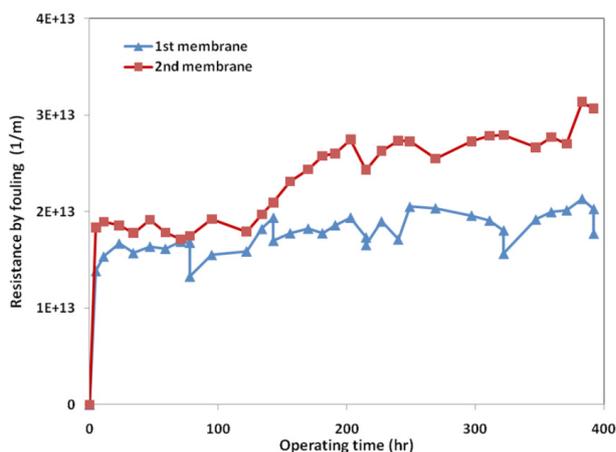


Fig. 10. Changes in filtration resistance by fouling with the passage of time (Conditions: TDS 30 g/L, operating pressure of 45 bar, injection of HA 50 mg/L+SA 50 mg/L).

in membranes in Series 2. According to Fig. 9, the concentration polarization factor also showed a similar tendency to the permeability coefficient. The concentration polarization factor showed a tendency to increase as high as the initial value. Applying the FOB in Series 1 alleviated reduction rate of concentration polarization factor and permeability coefficient. On the other hand, in Series 2, it continuously decreased by 14.21% compared to the initial value without FOB. Fig. 10 shows the filtration resistance by fouling using HA as foulant. Similarly, membrane fouling was effectively removed in Series 1 with FOB and lower than Series 2.

4. Conclusions

This study evaluated the concentration polarization and membrane fouling by calculating the permeation coefficient, concentration polarization factor and filtration resistance by fouling (R_f) with the progression of membrane fouling after forcibly injecting organic foulants at high concentra-

tions to accelerate membrane fouling. Moreover, this study evaluated the cleaning efficiency by analyzing changes in the permeation coefficient and concentration polarization factor with or without FOB and before and after FOB.

As fouling was progressed by hydrophilic organic matter (SA) and hydrophobic organic matter (HA), the permeation coefficient and concentration polarization factor decreased, and the filtration resistance by fouling increased. Fouling layers by HA and SA reduce the concentration polarization factor because they reduce the permeation coefficient by increasing the filtration resistance due to fouling and cause cake reduced concentration polarization by interrupting the convection of salt. Furthermore, membranes with FOB are effective in controlling membrane fouling because FOB slow down the decreasing rate of the permeation coefficient and concentration polarization factor and the increasing rate of filtration resistance by fouling compared to membranes without FOB.

Katsoufidou et al. [30] and Lee and Elimelech [29] reported that the flux recovery rate by physical cleaning was high because SA has high adhesion force between foulants but has low foulant-membrane interaction force. However, the increasing rate of the permeation coefficient and concentration polarization factor and the decreasing rate of the filtration resistance by fouling (R_f) measured in this experiment were higher on membranes fouled by HA. The results of this study are different from those of other studies. It was difficult to evaluate membrane fouling by comparing the permeability and concentration polarization coefficients of membranes fouled by SA, HA, and SA+HA and the filtration resistance by fouling. It was only possible to compare the efficiency with or without FOB in the same foulants.

Thus, it was possible to evaluate the degree of progression of membrane fouling in RO membranes by analyzing changes in the permeation coefficient and concentration polarization factor with the progression of membrane fouling or before and after FOB. This made it possible to use permeation coefficient and concentration polarization factor as an indicator to determine the timing of cleaning and physical cleaning such as FOB. Also, it is possible to use permeation coefficient and concentration polarization factor as an indicator to evaluate cleaning efficiency by analyzing the permeation coefficient and concentration polarization factor.

Acknowledgments

This research was supported by a grant (code 171FIP-C088924-04) from Industrial Facilities & Infrastructure Research Program funded by Ministry of Land, Infrastructure and Transport (MOLIT) of the Korea government and the Korea Agency for Infrastructure Technology Advancement (KAIA).

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