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Evaluation on cleaning efficiency and membrane fouling by permeation coefficient and concentration polarization factor in pilot-scale SWRO processes

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ABSTRACT

It is difficult to evaluate membrane fouling of seawater reverse osmosis (SWRO) membrane because fouling and concentration polarization occur at the same time. In particular, it becomes harder to distinguish the effects of fouling from those of osmotic pressure in full-scale SWRO membranes because six to eight elements are connected in series in one vessel and osmotic pressure increases more. Therefore, this study aimed to evaluate the applicability as an indicator of membrane fouling by monitoring permeation coefficient and concentration polarization factor with the progression of membrane fouling in a pilot plant with a capacity of 185 m^3/d operated by the constant flow method and by comparing them before and after maintenance chemical cleaning (MCC). Permeation coefficient and concentration polarization factor decreased with the progression of membrane fouling. However, after MCC, they increased because membrane foulants were removed by MCC. The cleaning efficiency of MCC by acid was superior to that by alkali because the increasing rates of permeation coefficient and concentration polarization factor in acid cleaning were higher than in alkaline cleaning. This is because the probability of forming scale by inorganic matter increased more than membrane fouling by organic matter without injecting antiscalants in order to realize chemical-free operation. In general, it is difficult to evaluate fouling because differential pressure (DP) monitoring RO membrane fouling did not change much. Conversely, the results showed that permeation coefficient and concentration polarization factor are highly applicable as a simple tool for evaluating membrane fouling because there was a clear tendency in fouling evaluation through them.

Keywords: Permeation coefficient; Concentration polarization factor; Fouling; Cleaning efficiency; Forward osmotic backwashing

1. Introduction

In general, membrane fouling occurs as a decrease in flux or trans-membrane pressure (TMP) [1–6], but it is difficult to monitor and evaluate membrane fouling in seawater reverse osmosis (SWRO) membranes [7–9]. The absence of changes in flux or TMP at the initial operation was regarded as a state without fouling because initially developed and commercialized SWRO membranes have high filtration resistance compared to low-pressure membranes [10,11].

However, because membrane resistance decreases and permeability increases in recently developed and commercialized SWRO membranes but fouling and concentration polarization occur at the same time due to the improvement of permeability, it is difficult to evaluate membrane fouling based on a decrease in flux and an increase in TMP [8,12–14]. With regard to actually used SWRO membranes, six to eight elements are connected in

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series in one vessel and osmotic pressure increases more because inlet feed water is concentrated through flow channels. As a result, it becomes more difficult to distinguish the effects of fouling from those of osmotic pressure [12,15–18].

Chong et al. conducted a study to distinguish concentration polarization from membrane fouling by spiking NaCl [19]. 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.

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

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

Song et al. reported that RO performance was controlled by thermodynamic equilibrium regions because osmotic pressure increased along flow channels in full-scale RO processes due to long channels [20]. In addition, they reported that RO 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 filtration resistance by fouling 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 [12]. I_f changes if F (F_1) 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 because they occur at the same time in SWRO processes. In particular, Nam et al. [21] evaluated membrane fouling by comparing and analyzing the permeation coefficient and concentration polarization factor for the lab-scale SWRO equipment operated using the constant-pressure method, but no study has been conducted to evaluate actual processes operated using the constant flow mode.

Therefore, this study aimed to evaluate the applicability as an indicator of membrane fouling by monitoring permeation coefficient and concentration polarization factor with the progression of membrane fouling in a pilot plant with a capacity of 185 m^3/d operated by the constant flow method and by comparing them before and after maintenance chemical cleaning (MCC).

2. Materials and methods

2.1. Experimental apparatus

The pilot plant is located in Udo desalination plant, Jeju City and has a capacity of 185 m^3/d. A schematic diagram of the process is shown in Fig. 1.

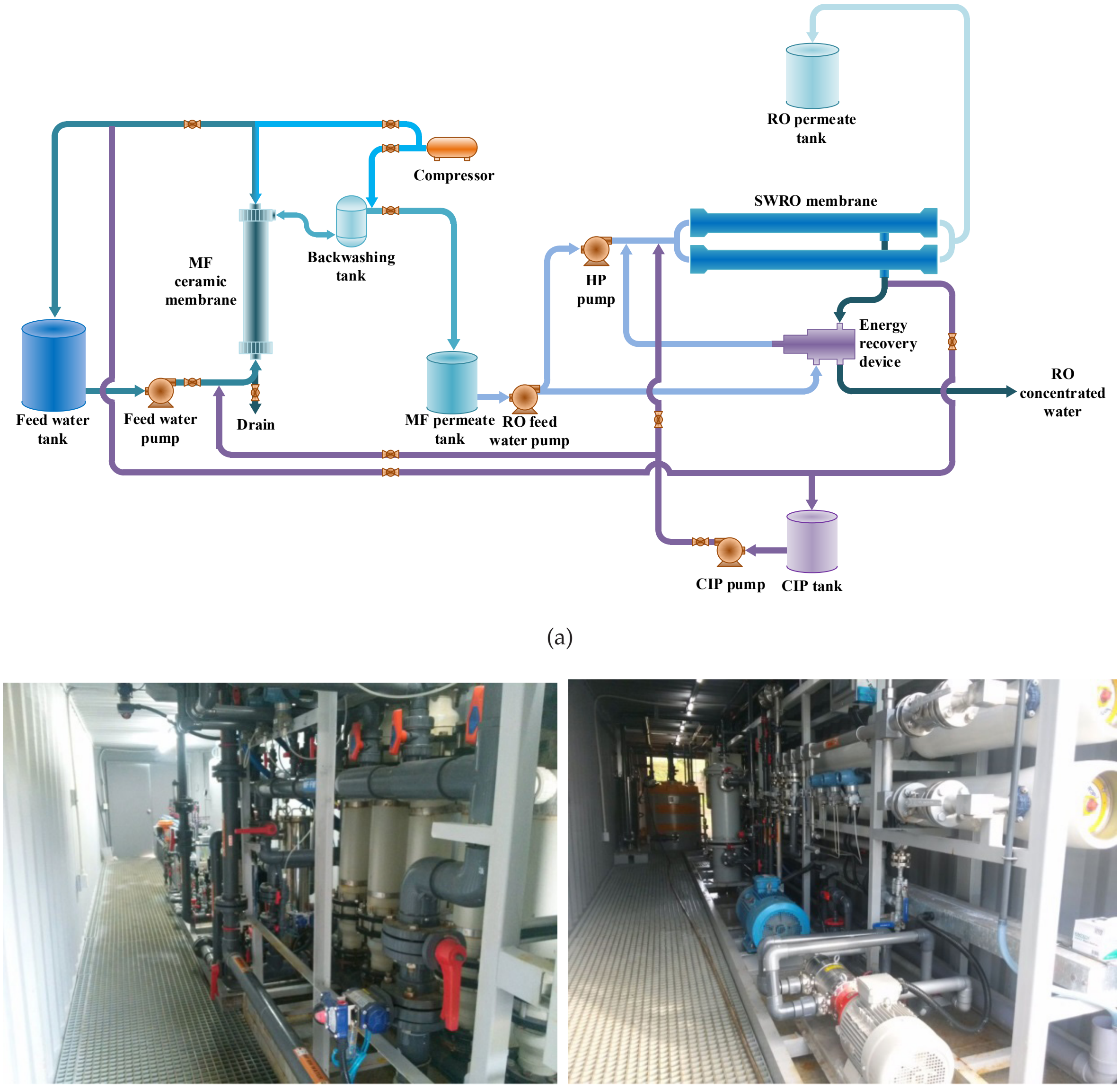
The pretreatment process for ceramic membrane filtration produces 370 m^3/d of treated water with a recovery rate of over 99%, and treated water flows into reverse osmosis (RO) membranes again (Fig. 1b). Residual chlorine was removed by injecting sodium bisulfate (SBS) before being flown to RO membranes because residual chlorine generated in chemically enhanced backwash of ceramic membranes causes damage to polyamide (PA) RO membranes. In addition, in order to improve the efficiency of the high-pressure pump, 185 m^3/day of the final treated water with a recovery rate of 50% was produced by installing an energy recovery device (ERD). Danfoss iSAVE, an isobaric type ERD, was installed.

With the recent development of high-flux membranes, permeate flux and fouling rate vary according to the position in one vessel. In general, the most severe fouling occurs in the membrane module located at the forefront end because it has the highest permeate flux and the lowest osmotic pressure. Therefore, in order to overcome this problem, hybrid module design to membrane module with high salt rejection rate at the front end of the vessel and high-flux membranes at the rear end is applied.

Peñate and García-Rodríguez simulated the membrane inter-stage design (HID) that several membrane manufacturers put different model membranes into one vessel to combine them. As a result, the design made it possible to reduce the use of vessel and energy costs because the design enabled operation at a high recovery rate and low operating pressure [22,23]. Thus, in order to increase the salt rejection rate and production volume, a total of two vessels were installed by filling one vessel on the inlet and outlet sides with five 400 SR membranes with high salt rejection rate and with two 400 ES membranes with high flux using the hybrid module design, respectively (Fig. 1c). A spiral wounded membrane made of thin-film nanocomposite produced in Nano H_2O were applied as an SWRO membrane. The detailed membrane specifications are shown in Table 1.

As shown in Fig. 2, when performing CIP according to the conditions suggested by the membrane manufacturer above CIP guidelines in the actual SWRO process, the targeting efficiency cannot be expected without increasing the cleaning concentration, extending the cleaning time, or enhancing the cleaning combination. Moreover, CIP is required with a shorter cycle because fouled membranes cannot be fully recovered and then fouling is accumulated. The cleaning efficiency gradually decreases, and eventually RO membranes should be replaced.

Thus, as shown in Fig. 3, there is a need for frequent MCC with low concentrations of chemicals in actual SWRO processes. This makes it possible to extend the chemical cleaning cycle and membrane lifetime by reducing membrane fouling, to increase accumulated production volume, and to reduce maintenance costs by decreasing electricity costs. In addition, antiscalants are injected as a pretreatment process to prevent scale formation because RO membranes



(a)

(b)

(c)

Fig. 1. Schematic diagram of pilot plant (a), pretreatment process for pilot plant (b), and primary treatment process for pilot plant (c).

are continuously operated. However, this study periodically performed MCC using NaOH, sulfuric acid, and EDTA to control scale without injecting antiscalants.

2.2. Experimental methods

The flux equation by the constant pressure mode in the study by Nam et al. was changed as shown in Eq. (1) by the constant flow method [21]. The horizontal and vertical axes correspond to feed water osmotic pressure and TMP, respectively. L_p and f_{cp} can be calculated because the gradient and y-section correspond to f_{cp}, y and J/L_p when floating with the data collected from the pilot plant.

$$\Delta P = f_{cp}\pi_b + J / L_p \tag{2}$$

where ΔP, f_{cp}, π_b, J, and L_p correspond to operating pressure in filtration, concentration polarization factor, average osmotic pressure of inlet water, water permeate flux, and membrane hydraulic permeation coefficient, respectively.

The feed water osmotic pressure was calculated using the empirical equation for seawater osmotic pressure as shown in Eq. (2), and this equation can be applied to the following conditions: from 10,000 to 80,000 mg/L at a temperature of 25°C [24]. The mean value of feed and concentrated water was used as TDS of inlet water applied to the calculation of osmotic pressure because one vessel in the pilot plant is filled with seven elements and they are concentrated with the progression of filtration [Eq. (3)].

$$\pi_b = 1.416\times10^{-7}C_a^2 + 6.913\times10^{-2}C_a - 80.64 \tag{3}$$

Table 1
SWRO membrane specifications

Parameter			Active area ft^2 (m^2)	Permeate flow rate gpd (m^3/d)	Stabilized salt rejection %	Max. applied pressure psig (bar)	Test condition				
							NaCl (mg/L)	Pres. psi (bar)	Temp. °F (°C)	Recovery (%.)	pH
SW RO	8″	Qfx SW 400 ES	400 (37)	13,700 (52)	99.8	1,200 (82.7)	32,000	800 (55)	77 (25)	8	8
		Qfx SW 400 SR	400 (37)	6,500 (24.6)	99.85	1,200 (82.7)					

SWRO membrane module (8″)

Actual installation in pilot plant

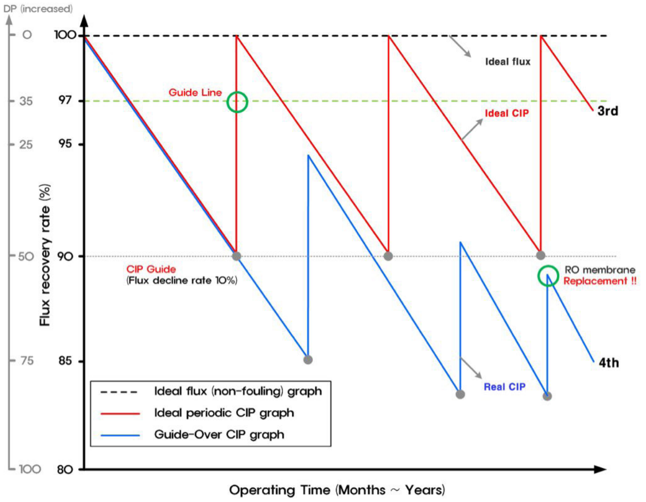


Fig. 2. Schematic diagram of chemical cleaning efficiency above guidelines for chemical cleaning.

$$C_a = \frac{C_f + C_b}{2} \tag{4}$$

where C_a, C_f, C_b, and π_b correspond to the mean inlet water TDS concentration (mg/L), feed water TDS concentration (mg/L), concentrated water TDS concentration (mg/L), and osmotic pressure (kPa), respectively.

This study aimed to evaluate how the permeation coefficient and concentration polarization factor changed with the progression of membrane fouling and how they were changed by MCC. During the experimental period, MCC was performed three times, and the cleaning conditions are shown in Table 5.3. MCC was performed by injecting NaOH in the first cleaning, sulfuric acid and EDTA in the second one, and only sulfuric acid in the third one. The cleaning cycle was gradually increased to 2, 3, and 4 weeks to evaluate the cleaning efficiency.

3. Results and discussion

3.1. Operating pressure and differential pressure

The operation results were summarized based on the data of the pilot plant operating between August and October in 2013. Inlet feed seawater was directly taken from a deep well, and TDS changes from 25 to 31 g/L twice daily according to tidal variation (Fig. 4). Fig. 4 shows the TDS of feed and treated water. TDS of RO treated water varies depending on TDS of inlet seawater because RO membranes showed a constant salt rejection rate of approximately 99%.

Fig. 5 is a graph showing changes in the inlet pressure of high-pressure pump into the pilot plant and DP in the whole operating time. The operating pressure varied changed from 50 to 60 bar in the reflection of changes in TDS depending on the tidal range. In general, DP monitoring RO membrane fouling did not change much despite the fact that fouling progressed with time. It also did not change much before and after MCC. These results show that it is difficult to evaluate fouling by DP and cleaning efficiency.

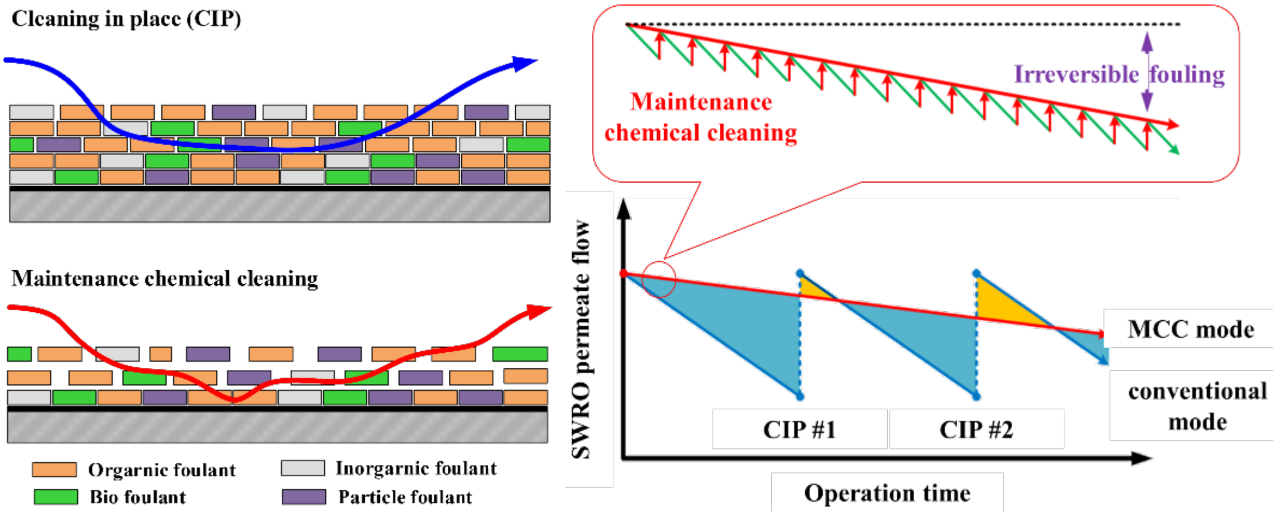


Fig. 3. Schematic diagram of MCC using chemicals in MCC.

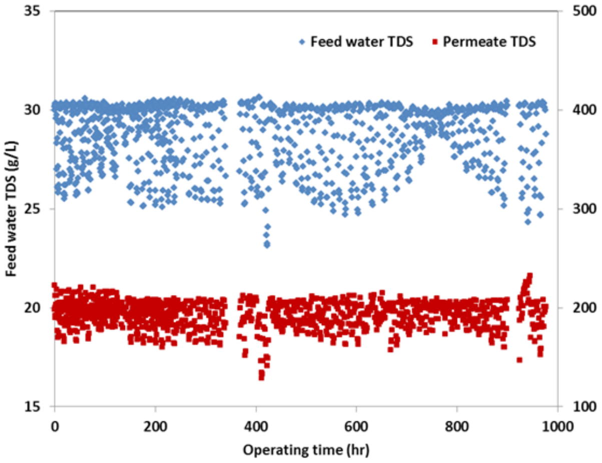


Fig. 4. Changes in TDS of inlet and treated water.

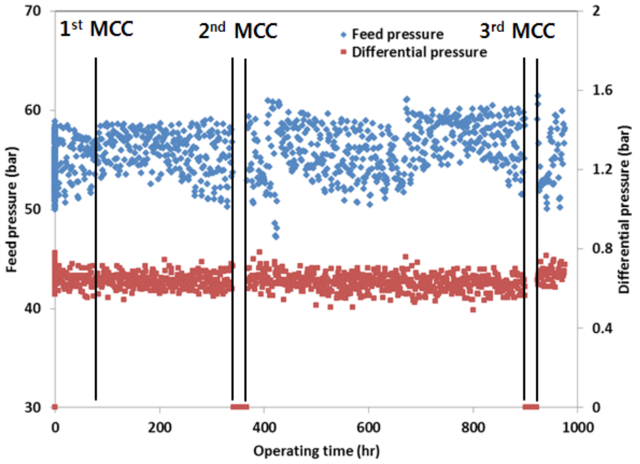


Fig. 5. Changes in operating pressure and differential pressure with time.

Table 2
Conditions for MCC

Session	Cleaning cycle	Cleaning condition
1st	2 weeks	NaOH pH 12 for 1 h
2nd	3 weeks	Sulfuric acid pH 3+EDTA 0.1% for 1 h
3rd	4 weeks	Sulfuric acid pH 3 for 1 h

3.2. Changes in permeation coefficient and concentration polarization factor

TDS varies according to the tidal range because inlet seawater is directly taken from a deep well. Therefore, the permeation coefficient and concentration polarization factor were derived by floating the results of osmotic pressure and inlet pressure when the TDS was the minimum value in the collected pilot data.

The results are shown in Fig. 6 and Table 3. According to Fig. 6, the permeation coefficient and concentration polarization factor show a tendency to decrease with the progression of membrane fouling. Immediately after the first MCC, the permeation coefficient decreased by 26.14% from 2.576 LMH/bar to 1.903 LMH/bar, and the concentration polarization factor decreased by 5.23% from 1.655 to 1.568. Moreover, after the second MCC, the permeation coefficient decreased by 38.17% from 2.370 LMH/bar to 1.466 LMH/bar, and the concentration polarization factor decreased by 9.29% from 1.665 to 1.511. The decreasing rate was relatively high because the cleaning cycle increased from 3 to 4 weeks.

The permeation coefficient decreased because the filtration resistance by fouling increased. The concentration polarization factor showed a similar tendency to the permeation coefficient and decreased with the progression of membrane fouling. 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 [25].

Furthermore, permeability coefficient and concentration polarization factor increased even in a constant flow

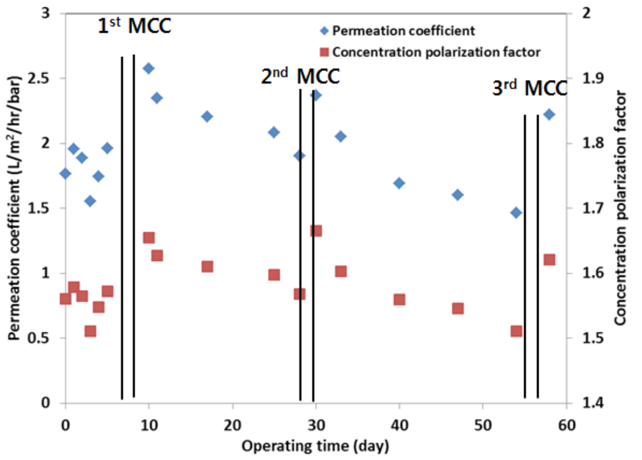


Fig. 6. Changes in permeation coefficient and concentration polarization factor with time.

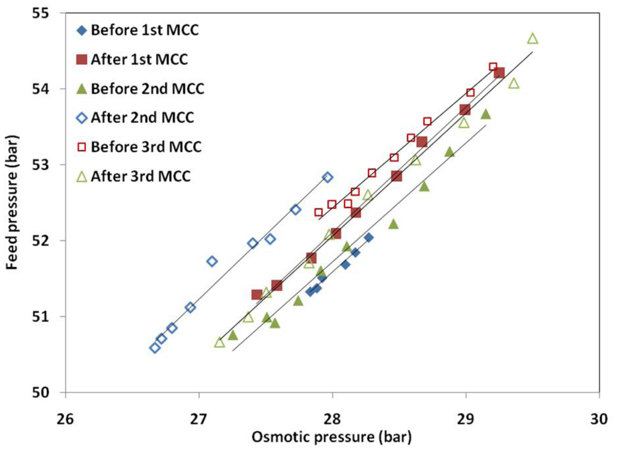


Fig. 7. Comparison of inlet pressure according to inlet water osmotic pressure before and after MCC.

Table 3
Changes in permeation coefficient and concentration polarization factor with time after MCC

Section	Number	Gradient	Y-section	L_p (LMH/bar)	f_{cp}
After 1st MCC	1	1.6548	5.7687	2.576	1.655
	2	1.6275	6.3346	2.346	1.627
	3	1.6104	6.7463	2.203	1.610
	4	1.5976	7.1450	2.080	1.598
	5	1.5683	7.8100	1.903	1.568
After 2nd MCC	1	1.6653	6.2693	2.370	1.665
	2	1.6026	7.2522	2.049	1.603
	3	1.5590	8.8006	1.689	1.559
	4	1.5458	9.2830	1.601	1.546
	5	1.5106	10.1390	1.466	1.511

rate filtration mode due to fouling occurrence. This showed a similar tendency to the decrease in permeability and concentration polarization coefficients with fouling operated in a constant pressure filtration mode.

The permeation coefficient and concentration polarization factor were plotted before and after MCC as shown in Fig. 7 and Table 4. The first, second and third MCC were performed under the conditions of Table 2 after operating for two, three, and four weeks, respectively. As shown in Table 4, before MCC for each condition, the permeation coefficient decreased to 1.963, 1.903, and 1.466 LMH/bar, and the concentration polarization factor also decreased to 1.572, 1.568, and 1.511. This was because fouling was progressed more with gradually increasing the cleaning cycle.

In addition, after MCC, the permeation coefficient increased by 31.22, 24.58, and 51.15%, and the concentration polarization factor also decreased to 5.27, 6.19, and 7.26%. The results showed that effective cleaning was performed. When comparing the cleaning efficiency, the first MCC is alkaline cleaning by NaOH, and the second and third MCC are acid cleaning by sulfuric acid. Acid cleaning shows relatively high cleaning efficiency. This is because the probability of forming membrane fouling, or scale, by inorganic matter increased more than membrane fouling by organic matter without injecting antiscalants as an RO pretreatment. Therefore, periodic MCC using acid solution makes it possible to perform operations even without injecting antiscalants and to extend the cycle of CIP. However, there is a need to derive the optimal condition by conducting cleaning experiments under various conditions such as cleaning cycle, cleaning concentration, and cleaning time.

4. Conclusions

This study evaluated membrane fouling and cleaning efficiency by deriving the permeation coefficient and concentration polarization factor based on operational data of the pilot plant operated in the constant flow mode with periodic FOB.

In general, it is difficult to evaluate fouling because DP monitoring RO membrane fouling did not change much. Conversely, the results showed that permeation coefficient and concentration polarization factor are highly applicable as a simple tool for evaluating membrane fouling because there was a clear tendency in fouling evaluation through them.

They decreased with the progression of membrane fouling. As result of applying MCC with different cycles such as two, three, and four weeks, the decreasing rates of permeation coefficient and concentration polarization factor increased and membrane fouling occurred more severely in proportion to the cycle. With the progression of membrane fouling, filtration resistance by membrane fouling increases, the permeation coefficient decreases, and the convection of salt was interrupted. Therefore, cake reduced concentration polarization (CRCP) was caused, and the concentration polarization factor decreases. The efficiency of MCC by acid was superior to that by alkali because the increasing rates of permeation coefficient and concentration polarization factor in acid cleaning were higher than in alkaline cleaning. This is because the probability of forming scale by inorganic matter increased more than membrane fouling by organic matter without inject-

Table 4
Comparison of permeation coefficient and concentration polarization factor before and after MCC

Section		Gradient	Y-section value	L_p(LMH/bar)		f_{cp}	
				Increasing rate (%)	Value	Increasing rate (%)	Value
1st	Before	1.5720	7.5694	1.963	31.22	1.572	5.27
MCC	After	1.6548	5.7687	2.576		1.655	
2nd	Before	1.5683	7.8100	1.903	24.58	1.568	6.19
MCC	After	1.6653	6.2693	2.370		1.665	
3rd	Before	1.5106	10.1390	1.466	51.51	1.511	7.26
MCC	After	1.6203	6.6918	2.221		1.620	

ing antiscalants in order to realize chemical-free operation. Furthermore, the permeation coefficient and concentration polarization factor increased because membrane foulants were removed by MCC.

Unlike low-pressure membranes such as microfiltration (MF) and ultrafiltration (UF), RO membranes are continuously operated without backwash, and it is difficult to monitor membrane fouling. Thus, CIP is performed with the shortened cycle in consideration of a safety factor. On the other hand, membrane fouling does not recover even if CIP is performed after operation above cleaning cycle on the assumption that membrane fouling did not occur because it is difficult to monitor membrane fouling.

Therefore, the introduction of permeation coefficient and concentration polarization factor is highly sensitive to makes it possible to determine the timing of maintenance chemical cleaning in reversible fouling. The CIP cycle can be increased and the amount of accumulated water can be maximized by determining the timing of appropriate cleaning because it is possible to evaluate fouling and cleaning efficiency more accurately. In addition, this makes it possible to save operating costs for chemicals and membrane replacement by extending the membrane lifetime.

Moreover, periodic MCC will be effective in improving operating performance such as reduction of electricity consumption of high-pressure pumps by reducing the increasing rate of RO membrane fouling and improvement of RO treated water quality and in ensuring the stability of processes.

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