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An evaluation on cleaning efficiency of forward osmotic backwashing based on organic matter analysis in SWRO processes

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ABSTRACT

Membrane fouling in seawater reverse osmosis (SWRO) occurs on the membrane surface to reduce the membrane performance. In particular, biofouling, silica scale, and calcium deposition are hardened with time and not easily removed because SWRO is continuously operated without shutdown. This study aimed to evaluate the performance of the SWRO membrane process by periodically applying forward osmotic backwashing (FOB) which is one of the physical cleaning methods. Dissolved organic carbon (DOC), UV_{254}, and fluorescence excitation-emission matrix (FEEM) were analyzed by sampling the circulating water before and after FOB. As a result, when sodium alginate (SA) as a hydrophilic substance was fouled, its cleaning efficiency was superior to that of humic acid (HA) as a hydrophobic material. This is because SA was effectively removed due to its low adhesion force between organic matter and membrane. In addition, FEEM of cleaning wastewater was analyzed after performing clean-in-place (CIP) of the membranes with and without FOB. The peak intensity with FOB was lower than that without FOB, and SA showed relatively large differences in the peak intensity with and without FOB. The effect of FOB on SA was larger than the cleaning efficiency for HA. In conclusion, FOB can extend the CIP cycle of SWRO. In particular, although the adhesion force between organic matter is high, FOB can mitigate membrane fouling by SA with small adhesion force with membranes.

Keywords: Reverse osmosis; Fouling; Forward osmosis backwashing; Cleaning efficiency

1. Introduction

Membrane fouling is a phenomenon whereby membrane resistance increases because foulants are deposited on the membrane surface or in the membrane [1]. In addition, membrane fouling in seawater reverse osmosis (SWRO) occurs on the membrane surface to reduce the membrane performance. In particular, biofouling, silica scale, and calcium deposition are hardened with time and not easily removed because SWRO is continuously operated without shutdown [2].

Thus, it is necessary to plan stable system operation by periodic cleaning. In general, cleaning is divided into cleaning-in-place (CIP) and physical cleaning. CIP is performed annually or biannually to remove foulants by reducing the adhesion between membrane and foulants using chemicals [3–9]. However, because CIP is performed with RO processes stopped, it may worsen the overall economic feasibility by reducing the production efficiency, increasing maintenance costs for using chemicals and treating cleaning wastewater, and advancing the timing of membrane replacement due to the deterioration of rejection performance, deformation and aging of membranes.

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On the other hand, physical cleaning methods include flushing, backwashing, vibration, and air sparging. Recently, forward osmotic backwashing (FOB) using osmotic pressure removes foulants attached to the membrane surface at the stage of reversible fouling, by running permeate to feed side. FOB has been actively studied because it is effective environmentally friendly technique [2,10–18]. Factors affecting the cleaning efficiency of FOB include feed water concentration (or circulated water concentration), operating pressure, and circulation flow rate. Among them, feed water concentration, or circulating water concentration, has the greatest impact, and operating pressure and circulation flow rate have a relatively small effect [2,11–13]. Moreover, factors affecting the cleaning efficiency include backwash flow rate, backwash cycle, and backwash time. Backwash flow rate and back wash cycle had a greater effect than backwash time.

Sagiv et al. reported that FOB performance was divided into two major steps such as a rapidly decreasing step by diluting the concentration polarization layer on the feed side and a gradually decreasing step [13]. In other words, concentration polarization (CP) layers are diluted with the progression of FOB, and the concentration on the membrane surface gradually decreases to become similar to the concentration of backwash water with the passage of time. Moreover, this study examined the effects of varying NaCl concentration, circulation flow rate, and operating pressure on accumulated backwash water volume to show the results graphically. Among them, feed water concentration, circulating water concentration, had the greatest impact, and operating pressure and circulation flow rate had a relatively small effect.

Nam et al. evaluated how total dissolved solids (TDS) of circulated water and backwash water affected the cleaning efficiency of FOB [19]. They prepared artificially circulated water and backwash water to apply FOB and evaluated the cleaning efficiency in comparison with the accumulated volume in FOB. As a result, the accumulated backwash volume increased with decreasing TDS of backwash water. However, although the initial backwash flow rate increases when TDS of circulating water are high, backwash flow rate rapidly decreases with the passage of time. In addition, cleaning efficiency decreases more with high TDS of backwash water than with low TDS of circulating water.

Treated water is applied as backwash water in real processes. Therefore, this study periodically applied FOB using actual treated water with the progression of membrane fouling. Moreover, this study aimed to evaluate the cleaning efficiency by comparing accumulated volume in FOB and to investigate the foulant removed by FOB by analyzing organic matter in concentrated water before and after FOB.

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.

Table 1
SWRO membrane specifications

Model	RE2521-SR
Effective membrane area, m^2	1.1
Permeateflow 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.

The used RO membrane experimental apparatus consisted of lab-scale cross-flow RO membrane test unit. The high-pressure pump, stirrer, water temperature controller, digital pressure gauge, and flow meter were linked to make possible automatic and continuous operation. The permeate flux 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 treated 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 tank. A chiller 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 flowmeter 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 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 natural organic matter (NOM) and extracellular polymeric substances (EPS), respectively. HA is a typical hydrophobic substance, and SA is a typical hydrophilic substance. 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 used HA and SA were supplied by Sigma-Aldrich. The organic matter concentration in the feed 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 running permeate to feed side using osmotic pressure and can be roughly divided into two methods: (1) backwashing by reducing the operating pressure to 0 or under the osmotic pressure [13,19], and (2) backwashing by generating osmotic pressure while maintaining operating pressure by injecting high concentration of salt [14]. In this study, FOB was performed by reducing the operating pressure to 0 using Method (1), and the operating pressure was rapidly reduced to 0 to minimize the effect of the conversion process in converting from filtration to FOB. FOB was conducted for 10 min, and the cleaning efficiency was evaluated by measuring the accumulated backwash volume in FOB [19,20].

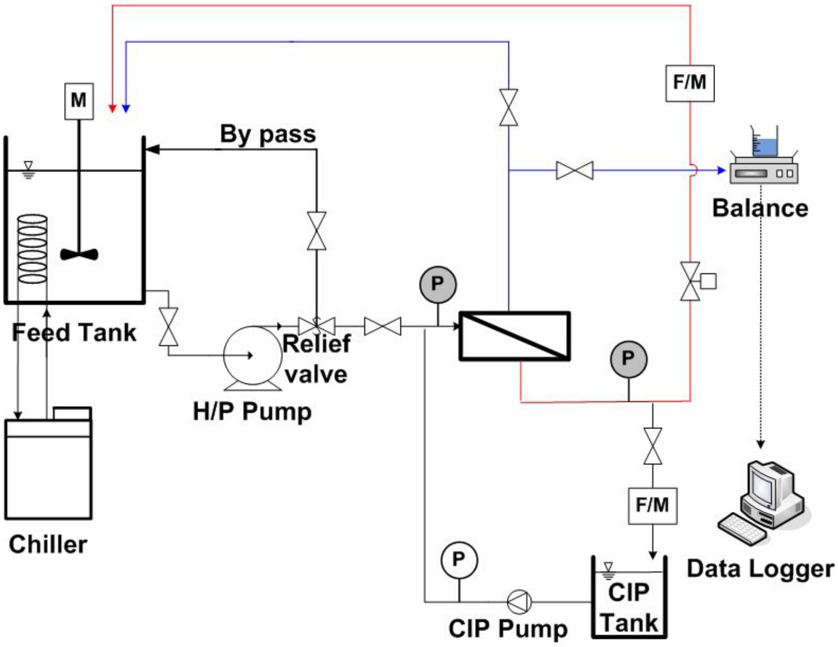


Fig. 1. Schematic diagram of experimental device for lab-scale continuous RO membrane.

In addition, this study aimed to investigate foulants removed on the membrane surface by FOB through UV_{254}, dissolved organic carbon (DOC), and fluorescence excitation-emission matrix (FEEM) analysis before and after FOB. HA and SA are evaluated by measuring UV_{254} and DOC, respectively [21]. They were analyzed using a UV-vis spectrophotometer (UVmini 1240, Shimadzu) and a total organic carbon (TOC) analyzer (TOC-V CPH, Shimadzu), respectively. FEEM was analyzed using a TECAN Safire 2.

3. Results and discussions

3.1. Evaluation of cleaning efficiency through accumulated backwash volume during FOB

Under fixed conditions at 30 g/L of TDS concentration and an operating pressure of 45 bar, FOB was performed periodically with the progression of fouling by injecting 50 mg/L sodium alginate of hydrophilic organic foulant and 50 mg/L HA of hydrophobic organic foulant. FOB was conducted five times for SA and six times for HA. The cleaning efficiency was evaluated by analyzing and comparing the accumulated volume before and after FOB. According to Park et al., the cleaning efficiency was analyzed by performing FOB under various conditions such as time, cycle, temperature of backwash water, and TDS in FOB. As a result, the cleaning efficiency increased with increasing accumulated volume [20].

With regard to the membranes fouled by SA and HA, Figs. 2 and 3 show the backwash flow rate and accumulated volume with the passage of time in FOB, respectively. As shown in Figs. 2 and 3, the flow rate and accumulated volume flowing into the feed side showed a similar tendency regardless of the type foulants. This agrees with the results of the modeling that Sagiv and Semiat divided FOB performance into a rapidly decreasing step by diluting the concentration polarization layers on the feed side (Step 1) and a gradually decreasing step (Step 2) [13].

Tables 2 and 3 show the features of membrane performance before and after FOB when injecting SA and HA. The water temperature during operation with the injection of SA was 23.8–23.9°C lower than 24.4–24.6°C which was the water temperature during operation with the injection of HA. This shows that membranes fouled by SA have a high salt rejection rate. In other words, the permeate water TDS of the membranes fouled by SA ranged from 552 to 570 mg/L lower than 760 to 821 mg/L which was the permeate water TDS of the membranes fouled by HA. However, permeate water TDS was more than 2,000 mg/L immediately after FOB. This is because internal concentration polarization (ICP) occurred in the membrane on the permeate side with the progression of FOB.

Factors affecting the backwash efficiency include feed water concentration(or circulated water concentration), operating pressure, and circulation flow rate. Among them, feed water concentration has the greatest impact. Although the driving force varies according to the feed water concentration, this is because TDS of permeate water produced by feed water also vary [2,12,13,20]. Permeate water was produced using the same feed water in this experiment, but TDS of permeate water vary because water temperature and foulants were different. TDS of this permeate water affected the accumulated backwash volume.

When comparing the final accumulated volume in Figs. 2 and 3 and Tables 2 and 3, the membrane fouled by SA with relatively low TDS of permeate water before FOB had a 20% higher accumulated FOB volume of permeate water. Nam et al. separately prepared TDS of circulating water and backwash water in FOB to compare the accumulated volume [19]. As a result, the accumulated backwash volume decreased with increasing TDS of backwash water in circulating water

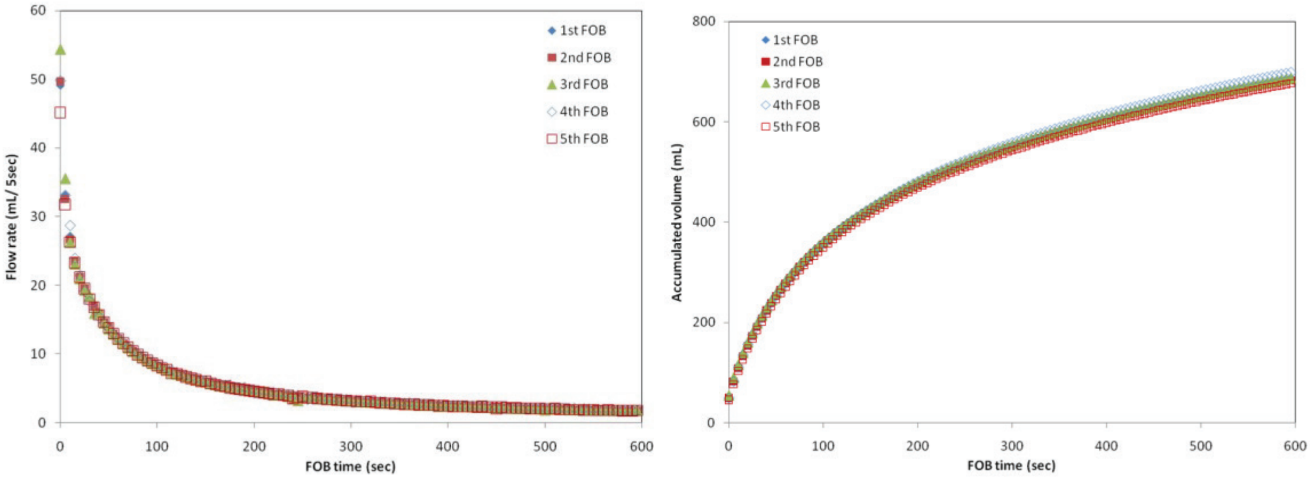


Fig. 2. Changes in flow rate (a) and accumulated backwash volume (b) in FOB of membranes fouled by SA.

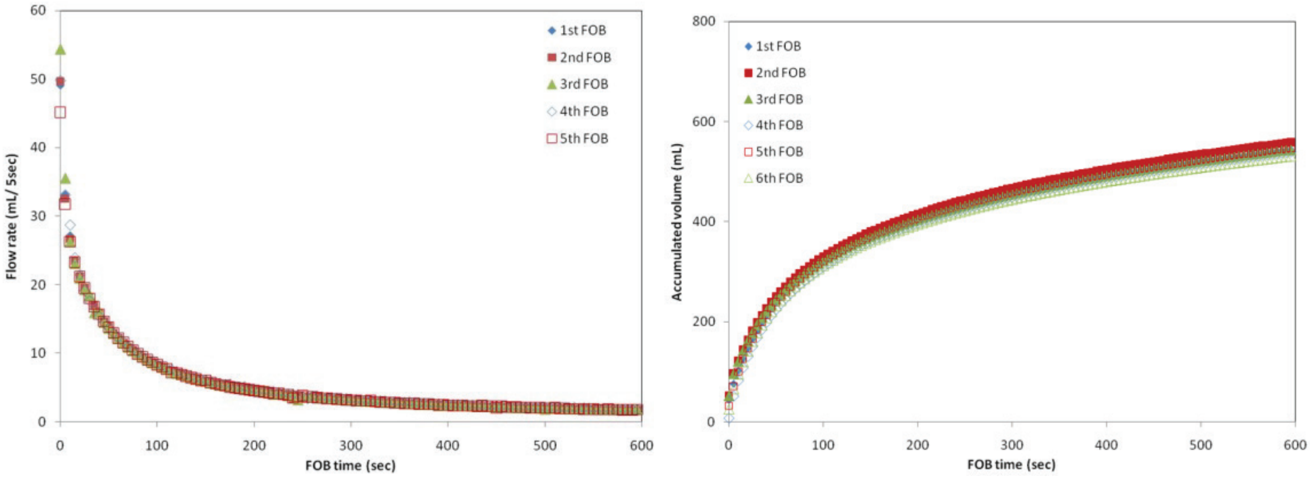


Fig. 3. Changes in flow rate (a) and accumulated backwash volume (b) in FOB of membranes fouled by HA.

Table 2
Characteristics before and after FOB on membranes with injection of SA

Session	Temperature (%)	Salt rejection rate (%)	Permeate r TDS (mg/L)		Accumulated volume of final FOB (mL)
			Before FOB	After FOB	
1	23.9	98.15	570	2,430	687.96
2	23.8	98.15	567	2,230	682.61
3	23.9	98.18	562	2,250	687.95
4	23.9	98.18	552	2,440	698.69
5	23.9	98.16	556	2,540	676.56

with the same TDS. Also, Katsoufidou et al. [21] and Lee and Elimelech [22] reported that the adhesion force between SA and SA is strong, but the interaction force between SA and membrane is weak. Therefore, FOB efficiency was increased during filtration due to interaction force.

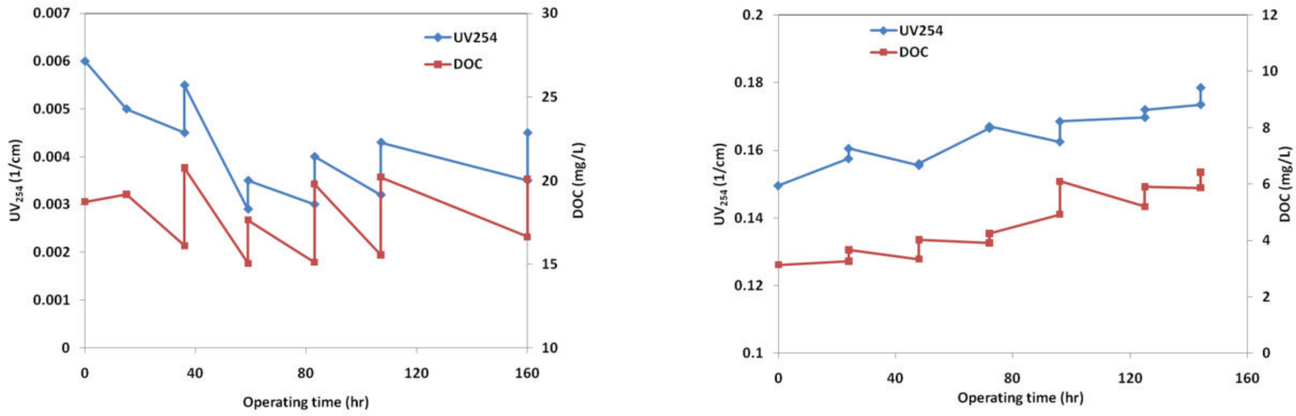
However, because the type of foulants and TDS concentrations of permeate water affected in these results at the same time, this study analyzed the organic concentration of concentrated water before and after FOB to identify whether they were affected by foulants.

3.2. Evaluation on the cleaning efficiency of FOB by organic matter analysis

This study evaluated the cleaning efficiency by measuring UV_{254}, DOC, and FEEM in the feed water tank

Table 3
Characteristics before and after FOB on membranes with injection of HA

Session	Temperature (%)	Salt rejection rate (%)	Permeate r TDS (mg/L)		Accumulated volume of final FOB (mL)
			Before FOB	After FOB	
1	24.6	97.47	767	2,300	555.84
2	24.5	97.44	760	2,120	558.84
3	24.4	97.31	806	2,870	545.22
4	24.4	97.35	816	2,950	540.28
5	24.4	97.26	794	2,860	547.34
6	24.4	97.28	821	2,960	530.76



(a) SA addition

(b) HA addtion

Fig. 4. Organic matter analysis of feed water with the passage of time by organic foulant.

before and after FOB and by comparing the volume of foulants removed from the membrane surface by FOB. In general, concentrations of HA and SA are evaluated by measuring UV_{254} and DOC, respectively [21]. The characteristics of SA and HA were compared by calculating the specific UV absorbance (SUVA) through the measurement of DOC and UV_{254}. SUVA is a key indicator of humic substances in the water, shows the degree of aromaticity, and is correlated with disinfection by-product precursors [23]. Conversely, with regard to membranes fouled by HA, UV_{254} ranged from 0.1556 to 0.1735 1/cm before FOB, but it increased by 0.30 to 3.69% to 0.1560 to 0.1720 1/cm after FOB. DOC ranged from 3.27 to 5.87 mg/L before FOB, but it increased by 8.52 to 23.75% to 3.67 to 6.42 mg/L after FOB. This is because Foulants are removed from the membrane surface when FOB is performed on fouled membranes, and then UV_{254} and DOC values increase. The increasing rate of DOC on membranes fouled by SA is higher than that of UV_{254} on membranes fouled by HA. Thus, it could be deduced that the cleaning efficiency by FOB is superior in hydrophilic organic matter. In particular, SUVA value, the ratio of UV_{254} and DOC, decreases with increasing hydrophilicity, but its hydrophilicity increased because it decreased on membranes fouled by SA+HA after FOB (figure omitted). In other words, this means that SUVA value decreased because much hydrophilic organic matter was removed from membranes than hydrophobic organic matter. This is because the cleaning efficiency was superior in hydrophilic organic matter and the increasing rate of DOC was larger increasing rate than that of UV_{254}. This agrees with results of Katsoufidou et al. [21] that the flux recovery rate was high after FOB because foulant-membrane interaction force was small. According to Katsoufidou et al., flux decline during filtering SA is not caused by foulant-membrane interaction force but caused by interaction force between foulants at the membrane fouled by SA [21]. In addition, this result shows a similar tendency to result of Lee and Elimelech [22].

The excitation/emission slit width of FEEM was 10 nm, and excitation/emission wavelength ranges were 230–800 nm and 280–800 nm. The actual peak intensity value was obtained from excitation/emission ranges of 230–700 nm/280–700 nm. On the basis of the values obtained by measuring the peak intensity of the excitation/emission wavelength combination obtaining the maximum fluorescence intensity from each specimen, characteristics of DOC were analyzed using the method developed by Chen et al. as shown in Fig. 5 [24]. According to Fig. 6, the excitation/emission peak intensity of feed seawater for SA and HA is mainly distributed in four areas and humic acid-like region, but intensity dis-

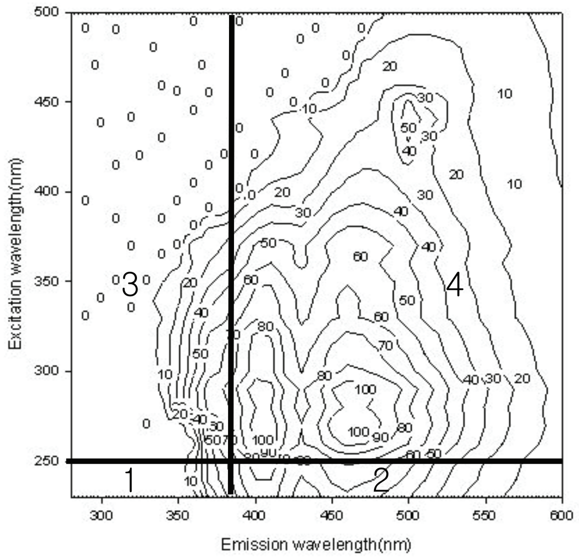
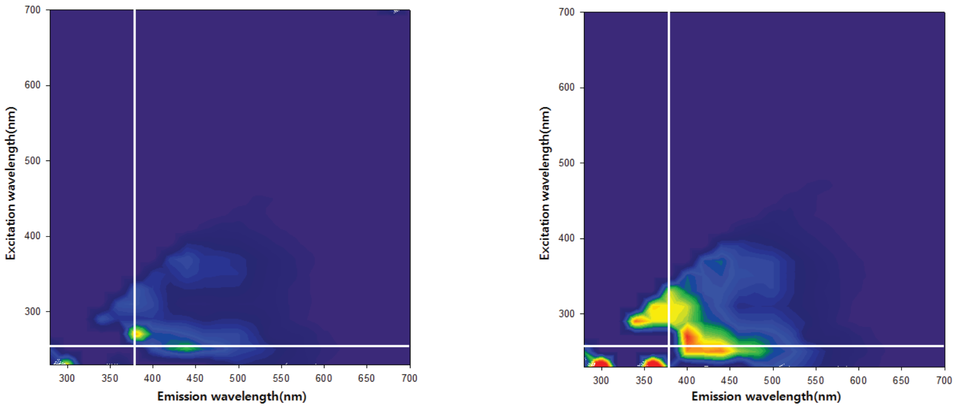


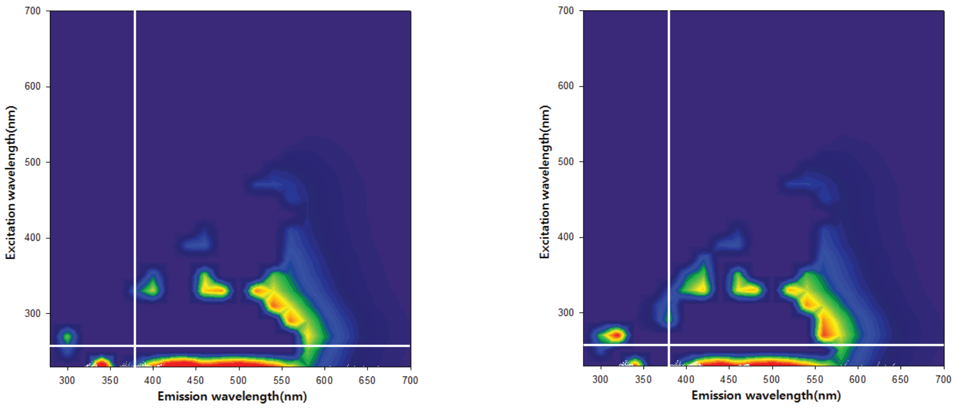
Fig. 5. Characteristics of organic matter for each FEEM fluorescent wavelength position; (1) Aromatic protein, (2) Fulvic acid-like, (3) Soluble microbial by product-like, (4) Humic acid-like.

tribution shapes were different from each other. Through comparing the intensity values before and after FOB, it could be deduced that foulants attached to the membrane surface were removed by FOB. Particularly, changes in the peak before and after FOB of membranes fouled by SA were larger than before and after FOB of membranes fouled by HA. This was because SA was attached to the membrane surface with low foulant-membrane interaction force before FOB and removed from the membrane surface more efficiently after FOB.

Fig. 7 shows the results of FEEM analysis of cleaning wastewater after chemical cleaning of membranes with and without FOB. NaOH at pH 12 was applied for two hours and peak intensity appears in a similar region to the peak intensity of feed water as the cleaning efficiency increased [25,26]. As shown in Figs. 7a, b comparing the intensity depending on the application of FOB for SA the peak intensity with FOB was slightly expressed in a narrower range than those without FOB. This is because FOB was effectively performed and a relatively small amount of foulants were attached to the membrane surface by FOB. On the other hand, as for HA, a similar tendency was observed with and without FOB because the effect of FOB was smaller than that in SA.

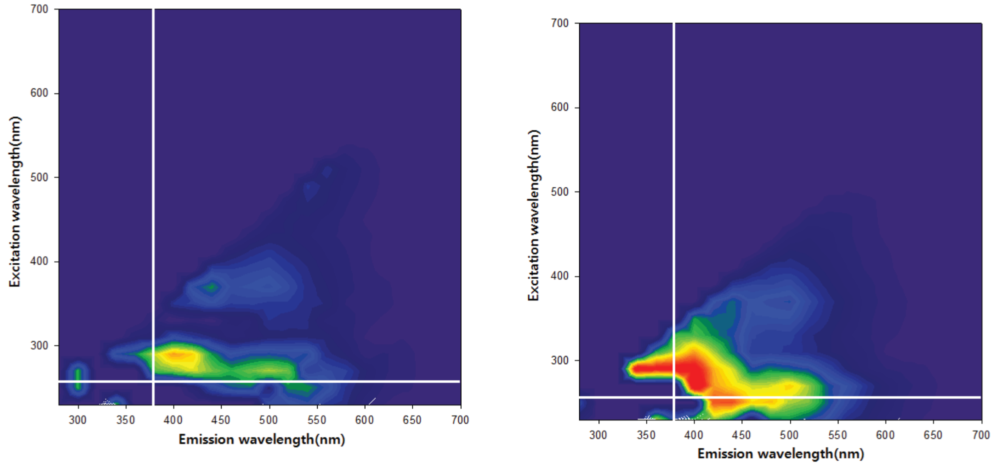


(a) Membranes fouled by SA before FOB (b) Membranes fouled by SA after FOB

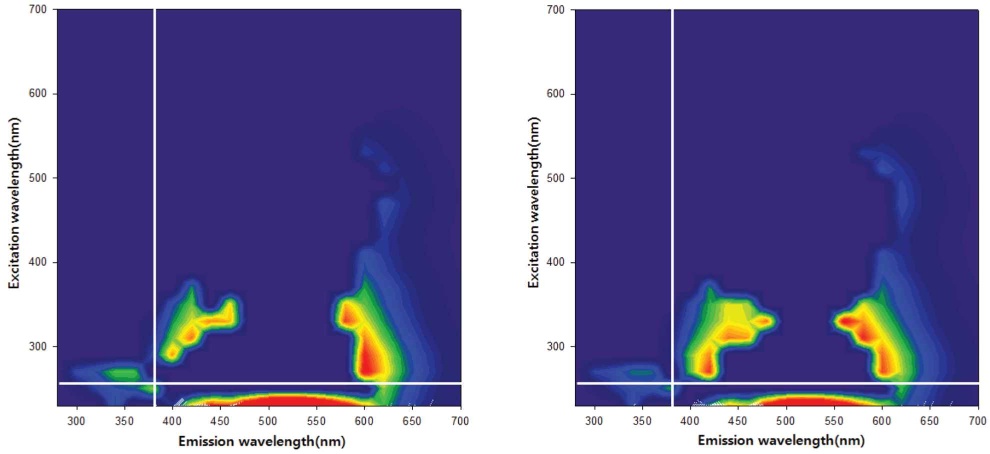


(c) Membranes fouled by HA before FOB (d) Membranes fouled by HA after FOB

Fig. 6. Comparison of FEEM before and after FOB of membranes fouled by SH and HA.



(a) Membranes fouled by SA (with FOB) (b) Membranes fouled by SA (without FOB)



(c) Membranes fouled by HA (with FOB) (d) Membranes fouled by HA (without FOB)

Fig. 7. Comparison of wastewater FEEM after CIP for membranes fouled by SH and HA.

4. Conclusions

This study periodically performed FOB using produced water as backwash water with the progression of membrane fouling for artificial seawater with organic foulants at high concentrations to accelerate membrane fouling and evaluated the efficiency of FOB by analyzing the accumulated volume and organic matter in the feed water tank during FOB.

The efficiency of FOB increased with increasing accumulated backwash volume. The cleaning efficiency was evaluated by the final accumulated backwash volume. As a result, it was considered that cleaning efficiency was superior because accumulated backwash volume increased with decreasing TDS concentration of backwash water when using treated water as backwash water. This is because internal concentration polarization (ICP) occurred in the membrane on the permeate water side with the progression of FOB. Therefore, in order to increase the cleaning efficiency of FOB, there is a need to consider water temperature, filtration time, and operating pressure affecting TDS concentration of treated water and to apply operating conditions capable of reducing the TDS concentration of treated water.

As a result of UV_{254}, DOC, and FEEM analysis before and after FOB, the cleaning efficiency was superior because the increasing rate of DOC on membranes fouled by SA is higher than that on membranes fouled by HA. In addition, FEEM analysis of cleaning wastewater after chemical cleaning of membranes with and without FOB was performed. The peak intensity of membranes with FOB was slightly expressed in a narrower range than membranes without FOB. This is because FOB was effectively performed and a relatively small amount of foulants were attached to the membrane surface. Conversely, as for HA, similar peak intensity was observed with and without FOB because the effect of FOB was smaller than that in SA. Therefore, it is expected that CIP cycles of SWRO membranes can be increased by periodically performing FOB to reduce membrane fouling by SA and then to operate SWRO plants effectively.

Acknowledgments

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