Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2018.22153

110 (2018) 19–26
April

Statistical optimization of chemical cleaning conditions of reverse osmosis membrane process in forward osmosis testing set-up

Pattarasiri Fagkaew, Yeo-Myeong Yun, Hyojeon Kim, Seoktae Kang*

Department of Civil and Environmental Engineering, Korea Advanced Institute of Science and Technology, 291, Daehak-ro, Yuseong-gu, Daejeon, 34141, Korea, Tel. +82 42 350 3635; Fax: +82 42 350 3610; email: stkang@kaist.ac.kr (S. Kang)

Received 19 December 2017; Accepted 25 January 2018

ABSTRACT

A chemical cleaning process is widely used to restore flux decline and solute rejection in membrane-based water treatment processes such as reverse osmosis (RO) process. In this study, the protocols for the chemical cleaning conditions in the RO process were optimized using a response surface methodology (RSM) for minimizing the experiment time and forward osmosis testing set-up, which can be operated with low or no hydraulic pressure, for reducing the usage of high hydraulic pressure pump. Fouled membranes were cleaned in accordance with the statistically designed conditions of various acid and alkaline agents with concentration, pH, and temperature as variables. Based on response surface plots, the chemical agent concentration was found to be the most influential factor to the membrane permeability. The optimum cleaning conditions obtained from RSM were 3% of the acid agent concentration at pH of 2.2 followed by 3% of the alkaline agent concentration at pH lower than 13. At these optimum conditions, the water flux was recovered about 86.1% with the salt rejection of 83.6%, compared with those of virgin membrane in RO process. Furthermore, this method could provide better understanding of the relationship among chemical cleaning agent concentration, pH, and temperature.

Keywords: Chemical cleaning; Reverse osmosis; Response surface methodology; Forward osmosis testing set-up

---

1. Introduction

Reverse osmosis (RO), a pressure-driven membrane process, has been widely used in the field of wastewater reclamation and desalination owing to its inherent advantages such as high contaminant rejection, simple operation, and low space requirement. However, a major impediment of the RO membrane process is fouling, which results in deterioration of membrane performance and thereby limiting the economic and technical feasibility of RO process. In the RO membrane process, the hydraulic pressure is applied to act as driving force for water permeation across a semi-permeable membrane. Meanwhile, the contaminants are rejected and formed as a layer on the membrane surface, called fouling layer [1]. In addition, the solute is also trapped in the fouling layer, leading to the significant cake-enhanced osmotic pressure and higher flux decline [2]. Numerous studies have been performed to develop strategies for governing and mitigating fouling, such as pre-treating feedstocks, improving the antifouling properties of membranes, and optimizing operating condition. However, despite these efforts, fouling is inevitable. Thus, frequent maintenance activities, such as chemical cleaning, are required to maintain the desired membrane performance.

The chemical cleaning is performed by dosing various chemical cleaning agents to remove the deposited foulants on the membrane surface by dissolving, hydrolyzing, or modifying the foulant structure or the fouling layer [3,4]. In general, chemical cleaning of membrane process is toward sequencing acid and alkaline agents to improve cleaning efficiency. This offers advantages of the maintained initial membrane performance, the decrease in operational costs, and the

---

* Corresponding author.

Presented at the 10th International Desalination Workshop (IDW 2017), November 22–25, 2017, Busan, Korea.

longer membrane life. In general, the chemical cleaning can be distinguished into two steps in accordance with the protocols in the RO process; an acid agent is used to remove inorganic precipitates by solubilization in the first step and an alkaline agent is used to remove organic matters by hydrolysis and solubilization in the second step [5]. Some additives such as surfactants are also added to the chemical cleaning agents to enhance the cleaning performance by forming micelles around foulants and solubilizing the macromolecules [6].

The chemical cleaning performance is effected by various factors such as chemical cleaning agent concentration, pH, and temperature. For a commercial membrane, it can be operated without threatening its integrity at the range of pH between 2.0 and 11.0 and operating temperature lower than 45°C. According to the previous study reported by Madaeni et al. [3], the cleaning efficiency increased with increase in the chemical cleaning agent concentration. However, the overdosing of the chemical cleaning agents can change the pH and lead to damaging membrane structure and decreasing the membrane lifespan. Temperature also plays important role in chemical cleaning process. When the temperature increases, it can cause the membrane swelling, resulting in the decrease in the membrane selectivity [7].

According to our previous study, the commercially available chemical cleaning agents designed for RO process could recover the reversal salt selectivity of fouled membrane up to 86% of initial flux after cleaning at company recommended operating conditions (1.5% of an acid agent concentration with a pH of 2.7 followed by 1.5% of an alkaline agent concentration with a pH of 11 at 35°C) in forward osmosis (FO) mode [8]. However, the optimization of that process has not been examined yet. Response surface methodology (RSM) has been very popular for optimization studies in recent years. This method is a combination of mathematical and statistical techniques which is useful for optimizing processes in which a response of interest is influenced by several variables [9]. It could help to minimize the load of experiments, resulting in the reduction of experiment time and cost. In addition, RO process requires high pressure pump to operate the system. To reduce the set-up installation as high pressure pump and stainless steel pipes, FO testing set-up which can be operated with low or no hydraulic pressure was conducted for performing the cleaning efficiency.

Based on this research background, the objective of this work was to optimize the protocols for chemical cleaning conditions in the RO process using RSM and FO testing set-up. The chemical cleaning agent concentration, pH, and temperature were selected as independent variables to optimize the cleaning conditions. The FO testing set-up was then used to determine the membrane permeability. Furthermore, water permeability and salt rejection also were investigated in the RO process to compare the cleaning efficiency. The changes of membrane surface properties including surface free energy, interfacial interaction, and Fourier transform infrared (FT-IR) spectra were measured to understand the recovery mechanism of the membrane performance.

2. Materials and methods

2.1. Preparation and chemical cleaning of membrane

A thin film composite RO membrane (RE-4040, CSM, Korea) was operated for 2 years for the water supply of steel industry in RO mode. It was mostly fouled with organic matter (77.7% mass of total foulants), which was analyzed by thermo-gravimetric analysis. The chemical components of foulants deposited on fouled membrane surface were examined by energy dispersive X-ray analysis; including C (74.72%), O (18.66%), and S (3.65%). Furthermore, it also included small amount of Al, Na, P, and Cl. The membrane was cut in pieces with 15 cm × 10 cm for the chemical cleaning and following FO tests. The chemical cleaning agents, including an acid agent and alkaline agent, were provided by RO cleaning chemical supplier (Prime Tech, Korea). The acid agent consists of peracetic acid and organic chelating acid, while the alkaline agent consists of two kinds of organic chelants with pH 10. Prepared fouled membrane pieces were then chemically cleaned with the acid agent followed by the alkaline agent in accordance with statistically designed conditions of various acid and alkaline agent concentration, pH, and temperature as shown in Table 1. All membrane samples were stored and soaked in deionized (DI) water at 20°C before using.

2.2. Design of chemical cleaning experiment

RSM with a central composite design, which provide lower number of experiments compared with normal full factorial design, was chosen and employed to determine the optimization of chemical cleaning with an acid and alkaline cleaning steps, respectively. Chemical cleaning agent concentration (X1), pH (X2), and temperature (X3) were selected as independent variables which were assessed at five coded levels (–1.682, –1, 0, +1, and +1.682), as shown in Table 1. In detail, the test A (acid cleaning step) was conducted to optimize the cleaning condition at the various acid concentration (0.3%–3.7%), pH (2.0–4.4), and temperature (18°C–42°C). Optimization of chemical cleaning with alkaline agent at the test B (alkaline cleaning step) was then performed at the various alkaline agent concentration (0.3%–3.7%), pH (8.6–11), and temperature (18°C–42°C). As a control, the membrane was cleaned by chemical agents according to the company instruction (an acid agent concentration of 1.5% and pH of 2.7 at 35°C followed by an alkaline agent concentration of 1.5% and pH of 11 at 35°C). The exposure time of each cleaning step was 12 h.

2.3. Flux recovery test

After chemical cleaning, the permeability (water flux and reverse salt flux) of membranes was tested for examining the relationship and comparing the cleaning efficiency in FO and RO modes. RO experiments were conducted in a laboratory scale RO cell unit, which was 15 cm in length and 10 cm in width (the total effective membrane area was 150 cm^2). 1,500 mg/L NaCl was used as feed solution at the pressure of 15 bar. For FO experiments, the experiments were conducted in a laboratory scale FO unit, which was 7.75 cm in length, 2.6 cm in width with a channel height of 0.30 cm (the total effective membrane area was 20.15 cm^2) [10]. DI water and 2 M NaCl (or 1.17×10^5 mg NaCl/L) were used as feed solution and draw solution, respectively. The flow of both sides was controlled by two variable speed pump (LongerPump WT3000-1FA, China) with a cross-flow velocity of 15 cm/s

Table 1
Design matrix of the RSM and experimental results

Trial no	Coded variables			Experimental variables						Test A (acid cleaning step)			Test B (alkaline cleaning step)		
				Test A (acid cleaning step)			Test B (alkaline cleaning step)								
	X1[a]	X2[b]	X3[c]	X1	X2	X3	X1	X2	X3	Jw[d]	Js[e]	RSS[f]	Jw	Js	RSS
1	–1	–1	–1	1.0	2.5	23	1.0	9.1	23	2.362	13.937	0.169	2.424	13.135	0.185
2	1	–1	–1	3.0	2.5	23	3.0	9.1	23	3.034	16.226	0.187	3.191	15.775	0.202
3	–1	1	–1	1.0	3.9	23	1.0	10.5	23	2.379	13.002	0.183	2.693	13.734	0.196
4	1	1	–1	3.0	3.9	23	3.0	10.5	23	2.993	14.868	0.201	3.081	15.174	0.203
5	–1	–1	1	1.0	2.5	37	1.0	9.1	37	2.384	13.682	0.174	2.492	13.417	0.186
6	1	–1	1	3.0	2.5	37	3.0	9.1	37	3.047	15.764	0.193	3.210	15.663	0.205
7	–1	1	1	1.0	3.9	37	1.0	10.5	37	2.460	14.700	0.167	2.950	14.306	0.206
8	1	1	1	3.0	3.9	37	3.0	10.5	37	3.069	16.026	0.192	3.195	15.723	0.203
9	–1.682	0	0	0.3	3.2	30	0.3	9.8	30	1.978	11.179	0.177	2.619	14.178	0.185
10	1.682	0	0	3.7	3.2	30	3.7	9.8	30	3.105	14.597	0.213	3.392	15.051	0.225
11	0	–1.682	0	2.0	2	30	2.0	8.6	30	2.521	14.002	0.180	2.671	15.456	0.173
12	0	1.682	0	2.0	4.4	30	2.0	11.0	30	2.503	13.281	0.188	2.983	15.211	0.196
13	0	0	–1.682	2.0	3.2	18	2.0	9.8	18	2.523	14.028	0.180	2.550	14.737	0.173
14	0	0	1.682	2.0	3.2	42	2.0	9.8	42	2.321	16.761	0.138	2.979	15.578	0.191
15[g]	0	0	0	2.0	3.2	30	2.0	9.8	30	2.333	14.202	0.164	2.828	14.066	0.201
16[g]	0	0	0	2.0	3.2	30	2.0	9.8	30	2.510	14.199	0.177	2.864	14.840	0.193
17[g]	0	0	0	2.0	3.2	30	2.0	9.8	30	2.237	15.595	0.143	2.785	14.495	0.192
18[g]	0	0	0	2.0	3.2	30	2.0	9.8	30	2.414	15.460	0.156	2.843	14.080	0.202
19[g]	0	0	0	2.0	3.2	30	2.0	9.8	30	2.724	14.173	0.192	2.709	14.171	0.191
20[g]	0	0	0	2.0	3.2	30	2.0	9.8	30	2.534	13.935	0.182	2.556	14.011	0.182

[a] Chemical cleaning agent concentration (%).
[b] pH.
[c] Temperature (°C).
[d] Water flux (L/m^{2}h).
[e] Reverse salt flux (mmol/m^2 h).
[f] Reversal salt selectivity (L/mmol).
[g] Center points.

for both sides. The water fluxes of both modes were determined by measuring the weight changes in the permeable side with a digital weight scale (A&D GF-4000, USA) which was connected to a computer during the entire experiment. The conductivity was also determined by the conductivity device (Vernier LabPro, USA). All experiments were carried out at 25°C ± 1°C.

2.4. Membrane surface analysis

Contact angles of membrane samples were measured by the contact angle analyzer (Phoenix SEO, Korea) using three liquids, including distilled water (W), ethylene glycol (E) and hexadecane (H). The surface free energy and interfacial interaction free energy was then calculated by Lifshitz-van der Waals/Lewis acid-base (LW/AB) method [11]. Furthermore, membrane morphologies were analyzed by scanning electron microscope (SEM) (Leica, Germany), and the functional groups on membrane surfaces were determined by Fourier-transformed infrared spectroscopy (FT-IR; PerkinElmer, USA).

3. Results and discussion

3.1. Relation of membrane permeability between RO and FO

To replace complicated RO tests with FO tests, the linear relation of membrane permeability between RO and FO mode should be proven. For that, the water flux and the reverse salt flux (or salt rejection in RO) of membrane samples, which were cleaned with different conditions, were plotted and then were compared in term of a correlation coefficient (*r*). If the magnitude of correlation coefficient is greater than 0.8, it is generally described as a strong linear relation, whereas the magnitude less than 0.5 is generally described as a weak linear relation [12,13].

The comparisons of the water flux and the salt flux between RO and FO modes are presented in Fig. 1. Both Figs. 1(a) and (b) showed that the correlation coefficients of

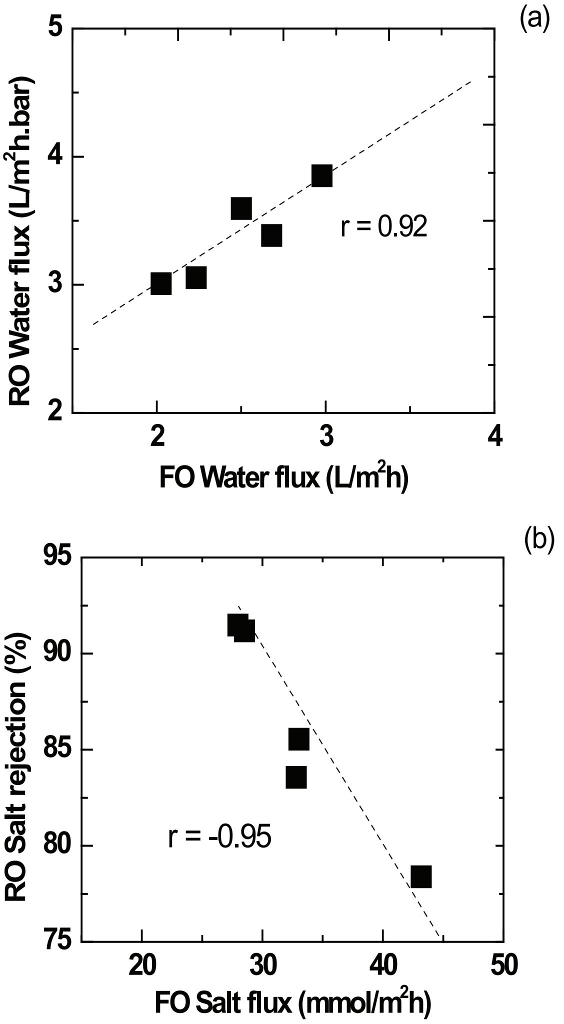

Fig. 1. Comparisons of (a) water flux and (b) salt flux (or salt rejection in RO) between RO and FO modes.

both modes were 0.92 and –0.95 for the water flux and the salt flux, respectively. It indicates that both water flux and salt flux (or salt rejection in RO) in RO and FO mode had the strong linear relation. Therefore, FO mode could be chosen to test the membrane permeability in RSM designed experiment due to its simple operation without applied hydraulic pressure pump, resulting in the decrease in set-up installation than operating in RO mode.

3.2. RSM application

3.2.1. Chemical cleaning step with an acid agent

Fouled membrane pieces were chemically cleaned with an acid agent as the first chemical cleaning step following the RSM designed conditions and then were tested in the FO testing set-up. The water flux, the reverse salt flux, and the reversal salt selectivity of each chemical cleaning condition were determined. They are shown in Table 1. To consider the influence of each operating factor on the membrane permeability, probability value (p-value) with 95% confidence level was determined. If p-value was less than or equal to 0.05 (p-value ≤ 0.05), it indicates that the factor significantly affects the membrane permeability [14]. According to RSM statistical results of water flux in the acid cleaning step obtained in FO mode as presented in Table 2, it could be seen that only the acid agent concentration (with p-value of 0.000) provided the p-value that was compatible to this condition. The acid agent concentration was determined as the most significant factor that affected the water flux with p-value of 0.000. The water flux increased with the increasing of the acid agent concentration. Meanwhile, pH and temperature were found to be less significant with p-value of 0.949 and 0.830, respectively. For the reverse salt flux, both acid agent concentration (with p-value 0.001) and temperature (with p-value of 0.040) provided the p-values that were compatible to this condition, so the acid agent concentration and temperature significantly affected to reverse salt flux. Reverse salt flux increased with the increase in acid agent concentration and temperature. However, the reversal salt selectivity, which indicates the performance of FO system, was mostly affected by the acid agent concentration with p-value of 0.024. Based on desirability function in RSM as shown in Figs. 2(a) and (b), the RSM optimum condition for high reversal salt selectivity were found as follows: the acid agent concentration of 3% and ambient pH after adding the acid agent at 30°C.

At the RSM optimum cleaning condition, it could recover the water flux of 82.1% with the reversal salt selectivity of 76.5% compared with the water flux and reversal salt selectivity of the virgin membrane, whereas the company recommended conditions only restored the water flux of 63.6% with reversal salt selectivity of 54.6% (Figs. 3(a) and (b)). It could be seen that the RSM optimum acid cleaning conditions could restore the membrane performance better than the company recommended conditions.

3.2.2. Chemical cleaning step with the alkaline agent

After cleaning fouled membranes at the RSM optimum acid cleaning conditions, the membranes were further cleaned with an alkaline agent as the second chemical cleaning step following the RSM designed conditions and then were tested in the FO testing set-up. The effect of operating factors on the water flux in the alkaline cleaning step, determining by p-value as can be seen in Table 2, found that all factors provided the p-value that were less than or equal to 0.05; the alkaline agent concentration (with p-value of 0.000), pH (with p-value of 0.008), and temperature (with p-value of 0.010). The water flux increased with the increase in these factors. On the other hand, the reverse salt flux was significantly affected by only alkaline agent concentration (with p-value of 0.001). It gradually increased with increase in the alkaline agent concentration. Whereas, pH and temperature were found to be less significant to the reverse salt flux with p-value of 0.798 and 0.214, respectively. For the reversal salt selectivity, the alkaline agent concentration was found to be the main factor affecting the reversal salt selectivity with p-value of 0.007. As shown in Figs. 2(c) and (d), the RSM optimum conditions for high reversal salt selectivity in the alkaline cleaning step were found as follows: the alkaline agent concentration of 3% at 30°C and controlled the pH lower than 13.0 due to the clean-in-place (CIP) pH range of this membrane was 1.0–13.0.

Table 2
Statistical results of membrane permeability in acid and alkaline cleaning step obtained from RSM

Factors	Test A (acid cleaning step)			Test B (alkaline cleaning step)		
	p-value (Jw)	*p*-value (Js)	*p*-value (RSS)	*p*-value (Jw)	*p*-value (Js)	*p*-value (RSS)
Concentration (%)	0.000	0.001	0.024	0.000	0.001	0.007
pH	0.949	0.454	0.542	0.010	0.798	0.053
Temperature (°C)	0.830	0.040	0.141	0.008	0.214	0.188

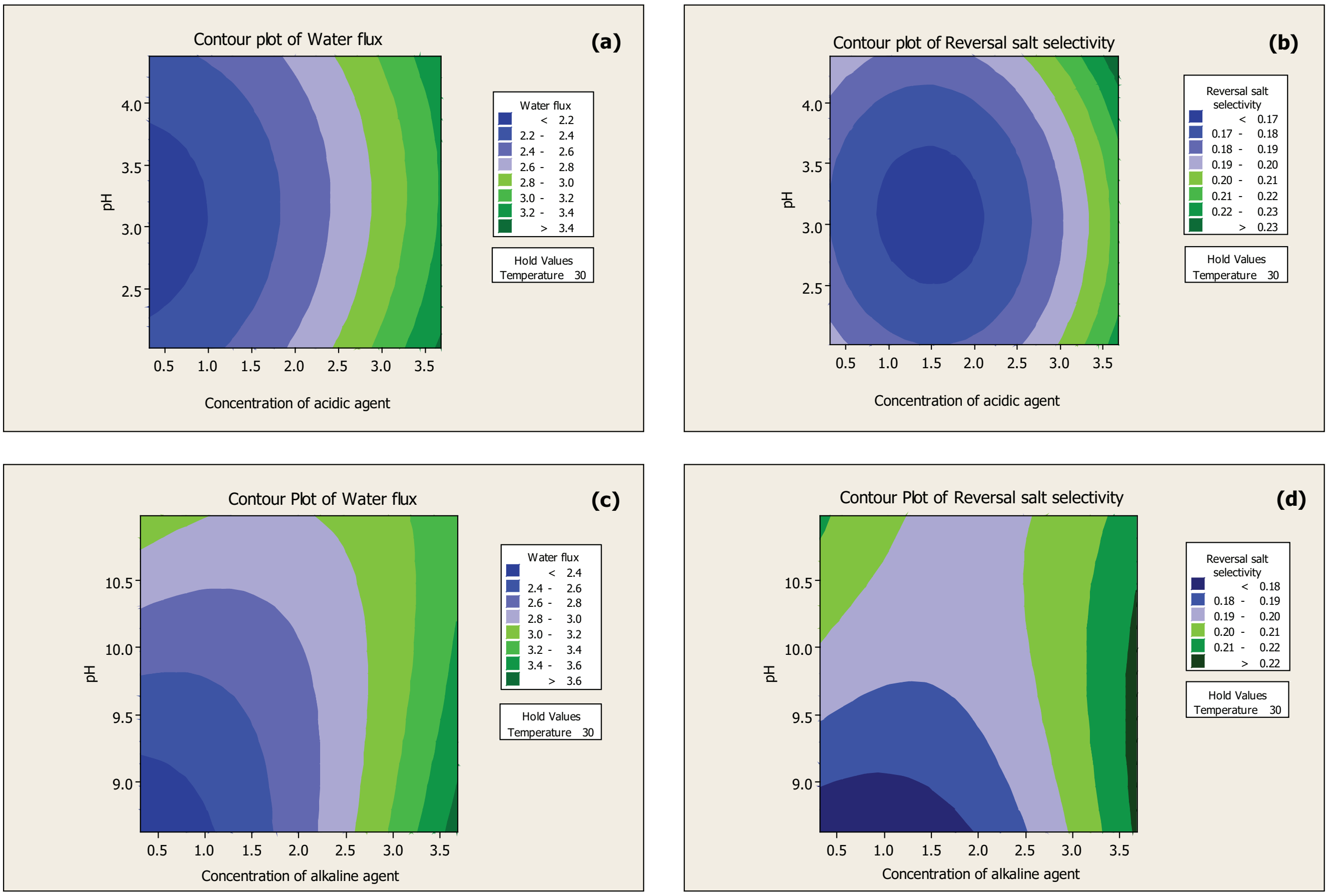

Fig. 2. Contour plots of water flux and reversal salt selectivity as function of operating factors in acid cleaning step ((a) and (b)) and alkaline cleaning step ((c) and (d)) at 30°C.

The alkaline cleaning step could also recover the membrane permeability as shown in Figs. 3(c) and (d). Cleaning fouled membrane with the optimum acid cleaning conditions followed by these optimum alkaline cleaning conditions recovered the water flux of 90.4% with the reversal salt selectivity of 83.7% compared with the virgin membrane. Meanwhile, the company recommended conditions only recovered the water flux of 78.9% with reversal salt selectivity of 60.6%. It could also be seen that the optimum cleaning conditions obtained from RSM could restore the membrane permeability better than the company recommended conditions in FO mode. In addition, the value of the reversal salt selectivity is independent of the structure of the membrane support layer, and solely determined by the selectivity of the membrane active layer [15]. Therefore, the increase of the reversal salt selectivity after the acid and alkaline cleaning steps were due to the recovery of membrane selectivity possible by the removal of fouling layers and recovery of surface properties.

To prove whether the suggested optimum conditions from RSM provides a better recovery, both membranes cleaned in RSM-optimum and company-suggested conditions were tested in RO mode with applied pressure of 15 bars at 25°C ± 1°C. 1,500 mg/L NaCl was used as feed solution through the RO experiment for 3 h. From Fig. 4, it can be confirmed that the RSM optimum cleaning conditions could recover the membrane permeability better than the company recommended conditions. The water flux was recovered

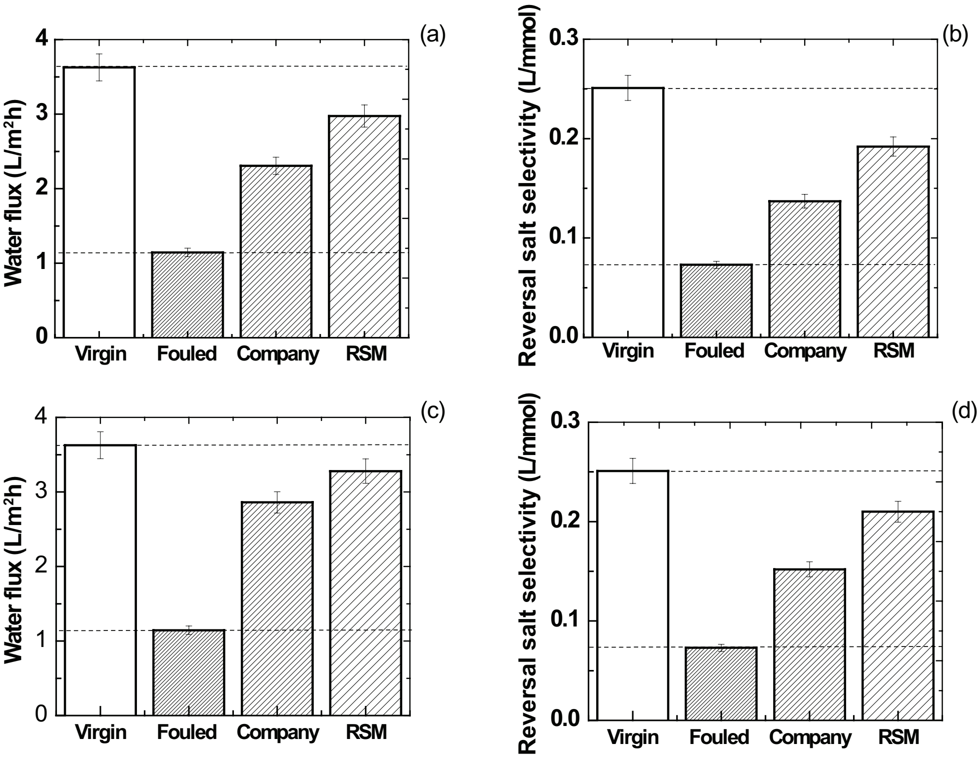

Fig. 3. Comparisons of water flux and reversal salt selectivity in acid cleaning step (a, b) and alkaline cleaning step (c, d).

about 86.1% with salt rejection of 83.6% at the optimum conditions, whereas the water flux of membrane cleaned with the company recommended conditions was only recovered 82.6% with salt rejection of 85.5%.

3.3. *Effect of membrane cleaning on membrane surface properties*

3.3.1. *Surface free energy and interfacial interaction free energy*

It is well known that the membrane permeability and fouling propensity of RO membrane is significantly affected by the membrane surface property such as hydrophilicity [16]. Generally, hydrophilicity was described in term of water contact angle. The lower water contact angle indicates the higher hydrophilicity [17]. However, the hydrophilicity of membrane surface in this work was presented in term of surface free energy (γ) and interfacial interaction free energy (ΔG_{MW}) which were calculated from the measured contact angles of membrane surface with two polar liquids (DI water, D and ethylene glycol, E) and a nonpolar liquid (hexadecane; H) using Lifshitz-van der Waal/Lewis acid-base method (LW/AB). Contact angle, surface free energy, and interfacial interaction free energy are shown and listed in Table 3.

According to Table 3, it can be seen that all four membrane samples exhibited high electron donating component (γ–) and low electron accepting component (γ+). The surface free energy of virgin membrane was 42.93 mJ/m^2, while that of fouled membrane was 29.04 mJ/m^2. Fuerstenau et al. [18] mentioned that the higher surface free energy can be indicated as higher hydrophilicity. It means that the decrease of surface free energy of virgin membrane after fouling was occurring due to the hydrophobic nature of foulants deposited on the membrane surface, changing the membrane surface became more hydrophobic. However, the surface free energy was recovered to 36.12 and 39.99 mJ/m^2 after cleaning with the company recommended and the RSM optimum conditions, respectively. The surface of cleaned membrane became more hydrophilic due to the removal of hydrophobic foulants (it could be further confirmed with FT-IR and SEM results). Moreover, the interfacial interaction free energy between membrane surface and water molecules, which indicated the favorable surface properties for the access of water molecules, also exhibited the similar trend. The larger negative value of ΔG_{MW} indicates the larger adhesive van der Waals force between membrane surface and water molecules, resulting in the higher water molecules transferring to membrane surface [19]. The interfacial interaction free energy of virgin membrane was –129.06 mJ/m^2, whereas that of fouled membrane was –95.63 mJ/m^2. After cleaning, the interfacial interaction free energy was decreased to –128.25 and –125.41 mJ/m^2 for the RSM optimum and the company recommended conditions, respectively. These results also correlated well with the water flux results. The water molecules could hardly approach to the fouled membrane surface, indicating the lowest water flux. In addition, the water molecules could easily approach to membrane which was cleaned with the RSM optimum conditions than membrane which was cleaned with the company recommended conditions, resulting in the higher water flux of membrane cleaned with the RSM optimum conditions. Therefore, the chemical cleaning with the RSM optimum conditions could recover the hydrophilic property of membrane with more acid-base interaction free energy, which indicated the more favorable surface properties for the access of water molecules than the company recommended conditions.

3.3.2. SEM and FT-IR

Images from SEM in Fig. 5 also confirmed that the surface of fouled membrane became more hydrophilic due to the removal of hydrophobic foulants after chemical cleaning. Fig. 5(a) presents the virgin membrane surface which was no foulants, whereas Fig. 5(b) shows the fouled membrane surface which was completely covered with foulants. After chemical cleaning with both conditions, SEM image showed the dramatic removal of foulants. However, the less noticeable amount of organic and colloidal matters was observed on the membrane surface which was cleaned with the RSM optimum conditions as shown in Fig. 5(d) than the membrane surface which was cleaned with the company recommended conditions (Fig. 5(c)).

Fourier transformed infrared spectroscopy (FT-IR) was also applied to confirm the removal of foulants of cleaned membrane in the range 450 to 4,000 cm^{-1}. From Fig. 6, the peaks of aromatic amines (1,240; 1,290; and 1,320 cm^{-1}), assigned to polyamide (PA) in the active layer [20] and polysulfonyl group (1,550 cm^{-1}) [21] in the porous polysulfone support layer which were observed in the virgin membrane surface, were disappeared on fouled membrane surface due to the deposition and formation of foulants layer. In addition, the fouled membrane displayed additional peak of 1,030 and 3,300 cm^{-1} which corresponding to the C–O stretching of carboxylic group and N–H stretching from amine group, respectively [22]. These peaks originated from an organic matter and/or microorganism deposited on the membrane surface. After chemical cleaning with both conditions, the peaks of PA were shown again and there were still small peaks of C–O and N–H stretching due to the residual organic and/or colloidal matters on membrane surfaces as shown in SEM results (Figs. 5(c) and (d)). It again confirmed the successful deposited foulants removal and recovery of original membrane properties due to chemical cleaning. Overall, FO mode

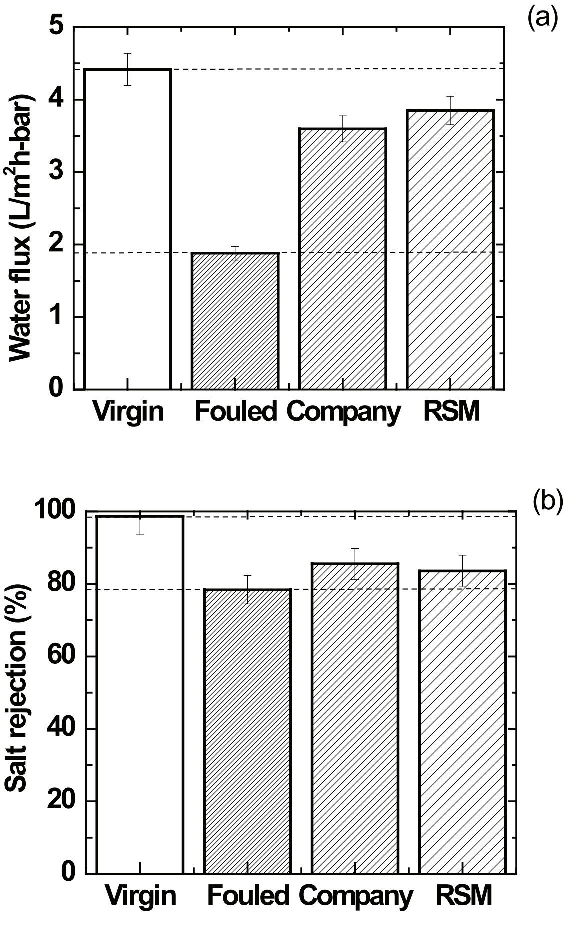

Fig. 4. Water flux (a) and salt rejection (b) of an organic fouled membrane sample operated in RO mode. Note that the initial flux was about 1.88 L/m^{2}h with 1,500 mg/L NaCl as feed solution.

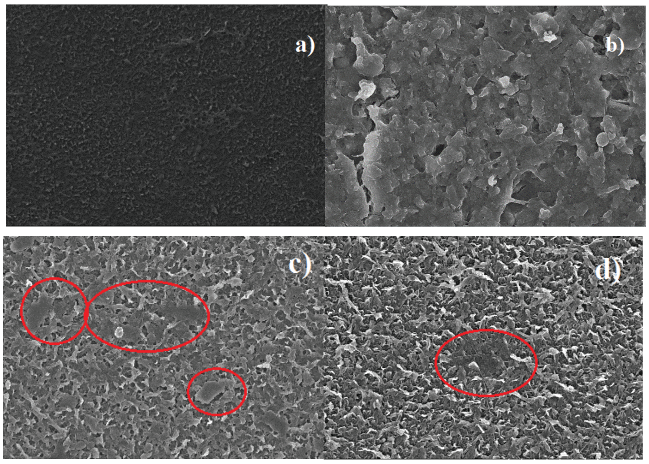

Fig. 5. SEM images of membrane samples (a) virgin, (b) fouled, (c) cleaned membrane with the company recommended conditions, and (d) cleaned membrane with the RSM optimum conditions.

Table 3
Contact angle, surface energy (mJ/m^2), and interfacial interaction free energy (mJ/m^2) of membrane samples

Samples	Contact angle			γ-LW	γ+	γ–	γ-total	ΔG_{MW}
	(W)	(E)	(H)					
Virgin	39.40	26.75	13.26	26.77	1.41	46.34	42.93	–129.06
Fouled	71.72	60.60	14.32	26.65	0.07	19.59	29.04	–95.63
Company	43.73	41.00	13.21	26.78	0.45	48.46	36.12	–125.41
RSM	40.39	33.47	14.19	26.67	0.91	48.54	39.99	–128.25

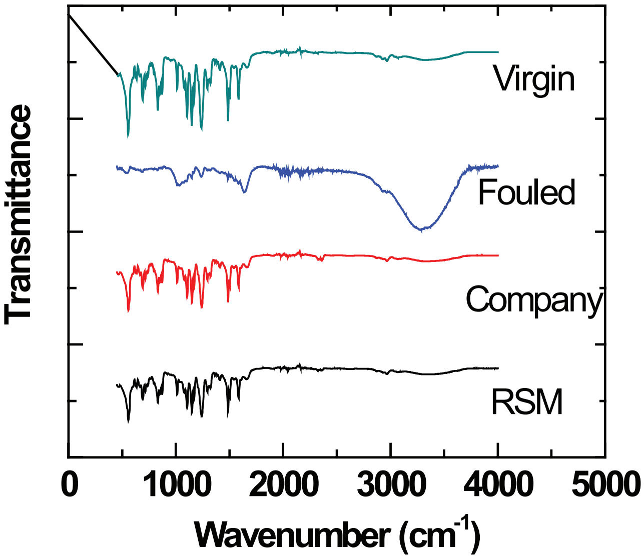

Fig. 6. FT-IR results.

coupled with a statistical analysis program was effective to be applied for determining the optimum chemical cleaning strategies in RO process. The RSM optimum chemical cleaning conditions could remove the foulants deposited on the RO membrane surface which changed the membrane surface became more hydrophilic and restored the higher membrane permeability than the company recommended conditions.

4. Conclusions

The objective of this study was to propose new statistical and experimental methods to determine the optimum regime for the chemical cleaning conditions in RO process. It was found that RSM coupled with FO testing set-up provided the better understanding between cleaning operational factors and membrane restoration efficiency. Moreover, it could be used as the fast and effective method to determine optimum cleaning strategies for chemical cleaning agents in the RO membrane process. The major factor affected to membrane performance was the concentration of cleaning agents, followed by pH and temperature. Based on desirability function in RSM, the optimum cleaning conditions obtained from RSM were 3% of the acid agent concentration at ambient pH after adding acid chemicals followed by 3% of an alkaline agent concentration at the pH lower than 13. These conditions could restore the water flux of 86.1% with salt rejection of 83.6%, compared with the virgin membrane in the RO mode, by improving more hydrophilic property of membrane surface than the company recommended conditions.

Acknowledgements

This research was supported by a grant (code 17IFIP-B088091-04) from Industrial Facilities & Infrastructure Research Program funded by Ministry of Land, Infrastructure and Transport of Korean government.

References

[1] L.F. Greenlee, D.F. Lawler, B.D. Freeman, B. Marrot, P. Moulin, Reverse osmosis desalination: Water sources, technology, and today's challenges, Water Res., 43 (2009) 2317–2348.
[2] S. Lee, C. Boo, M. Elimelech, S. Hong, Comparison of fouling behavior in forward osmosis (FO) and reverse osmosis (RO), J. Membr. Sci., 365 (2010) 34–39.
[3] S.S. Madaeni, T. Mohamamdi, M.K. Moghadam, Chemical cleaning of reverse osmosis membranes, Desalination, 134 (2001) 77–82.
[4] S.S. Madaeni, S. Samieirad, Chemical cleaning of reverse osmosis membrane fouled by wastewater, Desalination, 257 (2010) 80–86.
[5] J.M. Ochando-Pulido, M.D. Victor-Ortega, A. Martínez-Ferez, On the cleaning procedure of a hydrophilic reverse osmosis membrane fouled by secondary-treated olive mill wastewater, Chem. Eng. J., 260 (2015) 142–151.
[6] M.J. Rosen, D.S. Murphy, Synergism in binary mixtures of surfactants: VIII. Effect of the hydrocarbon in hydrocarbon/water systems, J. Colloid Interface Sci., 129 (1989) 208–216.
[7] A.J. Naaktgeboren, G.J. Snijders, J. Gons, Characterization of a new reverse osmosis composite membrane for industrial application, Desalination, 68 (1998) 223–242.
[8] K. Ruengruehan, P. Fagkaew, G. Ahn, S. Kang, S.-O. Ko, Relating membrane surface properties and flux recovery during the chemical cleaning of forward osmosis membrane, Desal. Wat. Treat., 57 (2016) 26621–26628.
[9] D. Baş, İ.H. Boyacı, Modeling and optimization I: Usability of response surface methodology, J. Food Eng., 78 (2007) 836–845.
[10] K. Ruengruehan, P. Fagkeaw, D. Jang, S. Ko, S. Kang, Impact of feed ionic concentration on colloidal and organic fouling of osmotically driven membrane process, Desal. Wat. Treat., 57 (2016) 24551–24556.
[11] T. Lin, Z. Lu, W. Chen, Interaction mechanisms and predictions on membrane fouling in an ultrafiltration system, using the XDLVO approach, J. Membr. Sci., 461 (2014) 49–58.
[12] R.M. Warner, Applied Statistics: From Bivariate Through Multivariate Techniques, SAGE Publications, 2012.
[13] M. Kurosu, Human-Computer Interaction: Interaction Modalities and Techniques, 15th International Conference, HCI International 2013, Las Vegas, NV, USA, July 21–26, 2013, Proceedings, Springer Berlin Heidelberg, 2013.
[14] L.V.A. Reddy, Y.-J. Wee, J.-S. Yun, H.-W. Ryu, Optimization of alkaline protease production by batch culture of *Bacillus* sp. RKY3 through Plackett–Burman and response surface methodological approaches, Bioresour. Technol., 99 (2008) 2242–2249.
[15] J.S. Yong, W.A. Phillip, M. Elimelech, Coupled reverse draw solute permeation and water flux in forward osmosis with neutral draw solutes, J. Membr. Sci., 392–393 (2012) 9–17.
[16] S. Azari, L. Zou, Using zwitterionic amino acid l-DOPA to modify the surface of thin film composite polyamide reverse osmosis membranes to increase their fouling resistance, J. Membr. Sci., 401–402 (2012) 68–75.
[17] T. Yasuda, T. Okuno, H. Yasuda, Contact angle of water on polymer surfaces, Langmuir, 10 (1994) 2435–2439.
[18] D.W. Fuerstenau, J. Diao, M.C. Williams, Characterization of the wettability of solid particles by film flotation 1. Experimental investigation, Colloids Surf., 60 (1991) 127–144.
[19] L. Feng, X. Li, G. Du, J. Chen, Adsorption and fouling characterization of *Klebsiella oxytoca* to microfiltration membranes, Process Biochem., 44 (2009) 1289–1292.
[20] R.M. Silverstein, F.X. Webster, D. Kiemle, D.L. Bryce, Spectrometric Identification of Organic Compounds, 8th ed., Wiley, 2014.
[21] Consultants Bureau Enterprises, Journal of Organic Chemistry of the USSR, Consultants Bureau, 1991.
[22] G. Socrates, Infrared and Raman Characteristic Group Frequencies: Tables and Charts, Wiley, 2004.