Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2016.1152650

57 (2016) 24532–24541
November

Taylor & Francis
Taylor & Francis Group

Comparison of fouling propensity and physical cleaning effect in forward osmosis, reverse osmosis, and membrane distillation

Yongsun Jang[a], Hyeongrak Cho[a], Yonghyun Shin[a], Yongjun Choi[a], Sangho Lee[a,*], Jaewuk Koo[a,b]

[a]*School of Civil and Environmental Engineering, Kookmin University, Jeongneung-Dong, Seongbuk-Gu, Seoul 136-702, Republic of Korea, Tel. +82 2 910 4529; Fax: +82 2 910 4939; emails: cutymonkey5@naver.com (Y. Jang), ggashigogi@naver.com (H. Cho), shinyonghyun@naver.com (Y. Shin), choiyj1041@gmail.com (Y. Choi), sanghlee@kookmin.ac.kr (S. Lee), koojaewuk@naver.com (J. Koo)*

[b]*Department of Environment and Plant Engineering, Korea Institute of Construction Technology, 2311 Daehwa-Dong, Ilsan-Gu, Goyang-Si, Gyeonggi-Do 411-712, South Korea*

Received 23 October 2015; Accepted 8 January 2016

ABSTRACT

Membrane-based desalination technologies including reverse osmosis (RO), forward osmosis (FO), and membrane distillation (MD) hold promise as efficient methods to produce fresh water from saline water sources. However, the fouling properties of these membranes are quite different due to the difference in the driving forces among them. Accordingly, the objective of this study is to compare fouling behavior and physical cleaning efficiency for these three membranes under similar operating conditions. Colloidal silica and alginate were used as model foulants and NaCl was added to feed solutions. Laboratory-scale experiments were carried out to compare fouling rates and recovery of flux by physical cleaning. Results showed that fouling propensity was the highest in FO membrane and the lowest in MD membrane, which may be attributed to the effect of cake-enhanced concentration polarization. On the other hand, physical cleaning was more efficient to recover flux in FO and RO than in MD, suggesting that the fouling in MD is less reversible than that in FO and RO.

Keywords: Desalination; Membrane distillation; Forward osmosis; Reverse osmosis; Membrane fouling; Physical cleaning

1. Introduction

The shortage of available water has become a critical issue around the world due to increasing populations and decreasing supplies of fresh water. Desalination of sea water can be one of the solutions to alleviate such problems [1]. The fresh water demand is growing and thus the worldwide desalination capacity is expected to expand continuously. Climate change by global warming will be a driver to accelerate the applications of desalination not only in arid areas but also in other countries [2].

*Corresponding author.

Presented at 2015 Academic Workshop for Desalination Technology held in the Institute for Far Eastern Studies Seoul, Korea, 23 October 2015

Many desalination plants adopt a technology called reverse osmosis (RO), which uses semipermeable polymeric membranes to separate dissolve ions from water. However, RO requires high energy to create a hydraulic pressure that exceeds the osmotic pressure of the saline solution. Due to this drawback, other technologies for desalination are being considered such as forward osmosis (FO) and membrane distillation (MD) [3]. FO is one of the evolving membrane technologies where a highly concentrated draw solution is placed opposite the feed solution to provide a driving force (osmotic pressure) to separate ions from water using a semipermeable membrane. [4–7]. FO can be used as a stand-alone system or a hybrid system combined with RO [8]. MD is another emerging membrane technology that uses vapor pressure as its driving force to produce fresh water from saline water [9,10]. Since MD is less sensitive to the osmotic pressure of feed solution, it can be used to concentrate high salinity feed water such as RO brine [11,12].

Although membrane technologies hold promise as efficient desalination methods, one of the annoying problems is membrane fouling, which leads to a decrease in separation efficiency and increase in energy costs to maintain its original productivity [13]. Fouling propensity of membrane systems depends on the properties of membrane materials, types of foulants, and the operating conditions such as flux and recovery. To develop effective strategies for fouling control, it is important to have a fundamental understanding of fouling behaviors and mechanisms [14,15].

There have been many studies on fouling behaviors of RO, FO, or MD membranes under various conditions [10,14,16–18]. Since the mechanisms of desalination by these membranes are quite different, different fouling phenomena have been reported. For example, it has been reported that fouling of FO membrane is less severe or more reversible than that of RO membrane [19]. However, few works have been attempted to simultaneously compare the fouling propensity of these membrane systems. Accordingly, this study intends to compare fouling propensity and physical cleaning effect of RO, FO, and MD membranes using same feed water conditions. Colloidal silica and alginate were selected as model foulants and background salt concentration was varied to examine the effect of ionic strength on membrane fouling. The rates of flux decline were compared as well as the reversibility of fouling by physical cleaning method, which helps to understand the different fouling mechanisms in different membrane systems.

2. Materials and methods

2.1. Feed solutions

The colloidal silica used as the model colloidal foulant is ST–ZL (SNOWTEX®, Nissan Chemical) with 70–100 nm particle sizes. This colloidal silica has 40 wt.% and pH is 9–10. Considering specific gravity (1.29–1.32), 5,000 ppm of silica feed solutions were prepared by dilution with deionized (DI) water. The alginate used as the model organic foulant is the medium viscosity alginic acid sodium salt from brown algae (Sigma–Aldrich). This alginate has $3.50 \pm 0.04 \times 10^5$ g/mol [20] and powder form. Alginate feed concentration used in fouling experiments was 500 mg/L

To identify the effect of salt concentration on fouling propensity by silica and alginate, NaCl was added to the feed solutions. NaCl concentrations were adjusted to 0, 1,000, and 35,000 ppm, respectively. The conditions of the feed water are summarized in Table. 1. Using these feed solutions, fouling tests for FO, RO, and MD membranes were carried out.

2.2. Experimental conditions

2.2.1. Forward osmosis

Fig. 1 shows the schematics of laboratory-scale FO membrane system used in this study. A commercial-grade polymeric FO membrane was used in the active layer (AL)-feed solution (FS) orientation. The length, width, and depth of the channel of the acrylic membrane module were 62, 15, and 2 mm, respectively and its effective membrane area was 12.5 cm^2. A micro gear pump was used to supply feed and draw solution at 0.5 L/min, which corresponds to crossflow velocity of 0.19 m/s. In all FO fouling experiments, volume of feed and draw solutions was 1 L. The operation of FO was carried out in a counter-current flow configuration. The NaCl solution of 4 M was used as

Table 1
Feed conditions according to the type of foulants and foulant concentrations

Division of feed conditions	
Feed 1	Silica 5,000 ppm
Feed 2	Silica 5,000 ppm and NaCl 1,000 ppm
Feed 3	Silica 5,000 ppm and NaCl 35,000 ppm
Feed 4	Alginate 500 ppm
Feed 5	Alginate 500 ppm and NaCl 1,000 ppm
Feed 6	Alginate 500 ppm and NaCl 35,000 ppm

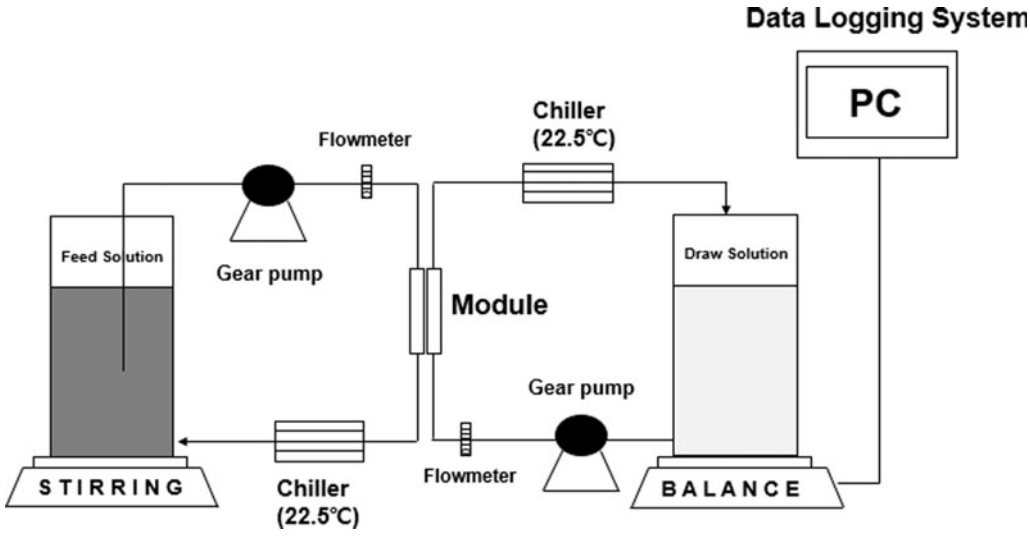

Fig. 1. Schematic diagram of laboratory-scale FO system.

the draw solution. Temperature of feed and draw solution was maintained at 22.5°C using a chiller. An electronic balance was connected with PC to continuously measure water flux.

2.2.2. Reverse osmosis

The schematic diagram of laboratory-scale RO membrane system used is shown in Fig. 2. A thin film composite RO membrane was used in this study, which was SW30-2521 (DOW FILMTEC™) modules. The water permeability of this membrane was experimentally determined at 1.6 LMH/bar using a dead-end stirred cell. The length, width, and depth of the channel of the laboratory-scale RO membrane module were 86, 49, and 1 mm, respectively. The effective membrane area was 43 cm^2 and the flow rate was 0.5 L/M, which corresponds to crossflow velocity of 0.17 m/s. During the RO runs, feed temperature was kept constant at 22.5°C. The feed volume in RO process was 3.5 L because the effective area of the RO membrane was about 3.5 times larger than that of the FO and MD membranes. An additional RO experiment was carried out using NaCl 35,000 ppm solution without any foulant.

2.2.3. Membrane distillation

Fig. 3 illustrates the schematic diagram of the laboratory-scale MD system. In the MD tests, a

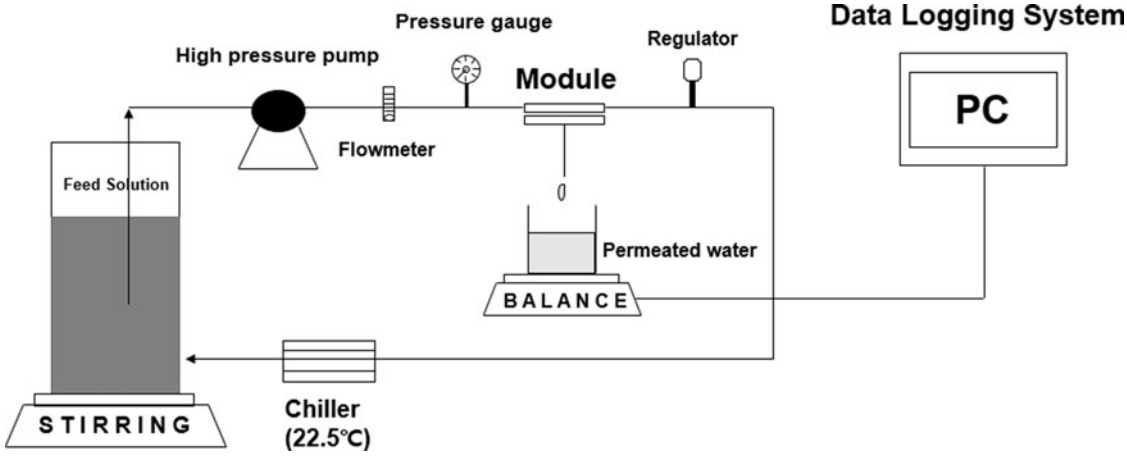

Fig. 2. Schematic diagram of laboratory-scale RO system.

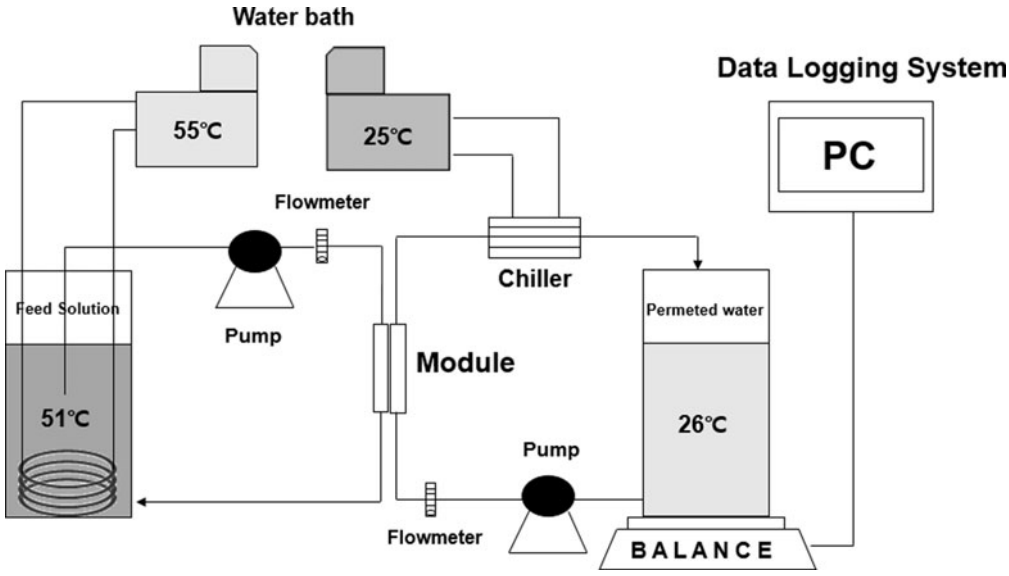

Fig. 3. Schematic diagram of laboratory-scale DCMD system.

hydrophobic PTFE flat sheet MF membrane (Millipore, USA) was used, which has the nominal pore size of 0.22 μm pore. The dimension of MD module was same as that of the FO module. The direct contact MD (DCMD) configuration was selected. The temperatures of the feed solution and permeate were 51 and 26˚C, respectively. The operation of MD was also carried out in a counter-current flow configuration. The volume of feed and permeate water was 1 L.

3. Results and discussion

3.1. Fouling by colloidal foulants (silica)

The changes in flux in FO, RO, and MD membranes with time were compared using feed solutions containing the colloidal silica. The results are shown in Fig. 4. No significant flux decline was observed when the RO and MD membranes were applied to treat feed solution containing only colloidal silica. On the other hand, rapid flux loss occurred when the FO membrane was used, as shown in Fig. 4(a). Within 500 min, the FO flux was reduced by 70%. Increasing the NaCl concentration up to 1,000 mg/L did not affect the fouling behaviors of these membranes, as shown in Fig. 4(b). However, fouling was detected in all three membranes when the feed contained the colloidal silica of 5,000 mg/L and NaCl of 35,000 mg/L as depicted in Fig. 4(c). The relative values of flux after 500 min for the FO and RO membranes were only 0.3 and that for the MD membrane was approximately 0.45, which is summarized in Fig. 4(d).

The effect of salt concentration in the feed solution on flux decline in FO system was investigated to understand the fouling mechanisms of FO membrane. As shown in Fig. 5, the normalized flux (the ratio of final flux to initial flux) increased with increasing the NaCl concentration in the feed solution. This can be explained by introducing the concept of the cake-enhanced osmotic pressure (CEOP) phenomena by the reversed solutes flux (RSF). According to this mechanism, the cake layer hinders the back diffusion of salt passing from membrane surface to the bulk solution, resulting in elevation of osmotic pressure near the membrane surface and severe flux decline [21–25]. Accordingly, it is likely that significant flux drop in FO experiments results from CEOP caused by RSF. As the NaCl concentration in the feed solution increases, the RSF may be decreased, leading to less severe CEOP effect.

On the other hand, the CEOP in RO system is caused by the ions in the feed solution [21] because there is no RSF in RO system. Accordingly, fouling occurred by the feed solution containing colloidal particles under high ionic strength while no fouling occurred under low ionic strength conditions, as shown in Fig. 4(c). Similar phenomena seem to occur in MD system, leading to flux decline in the presence of cake layer under high ionic strength. Nevertheless, the normalized flux for MD is higher (0.45) than that of FO and RO (0.3) under these conditions, suggesting that MD is less sensitive to flux decline by CEOP. This is probably because MD is not significantly affected by concentration polarization phenomenon [9].

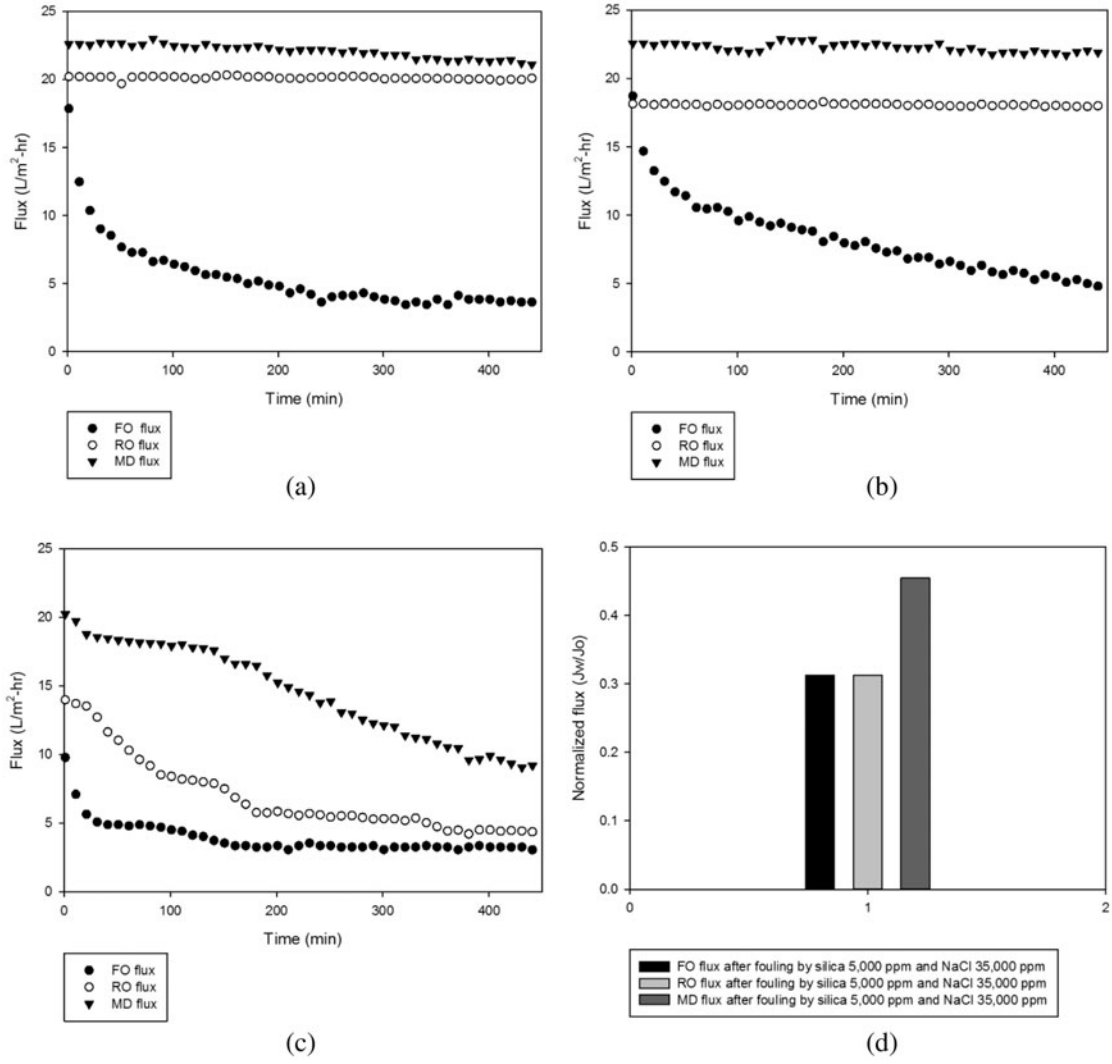

Fig. 4. Comparison of flux behaviors in FO, RO, and MD systems: (a) Feed water containing colloidal silica of 5,000 mg/L, (b) Feed water containing colloidal silica of 5,000 mg/L and NaCl of 1,000 mg/L, (c) Feed water containing colloidal silica of 5,000 mg/L and NaCl of 35,000 mg/L, and (d) Normalized flux after the experiment.

3.2. Fouling by organic matters (alginate)

In Fig. 6, the dependence of flux in FO, RO, and MD membranes on time was compared using feed solutions containing alginate. Again, no significant flux decline was observed in RO and MD systems using the feed solution containing only alginate. On the other hand, a rapid flux drop occurred in FO system, as depicted in Fig. 6(a). Increasing the NaCl concentration up to 1,000 mg/L does not affect the flux behaviors, as shown in Fig. 6(b). However, the flux in RO system was reduced from the beginning when the feed water containing alginate of 500 mg/L and NaCl of 35,000 mg/L was used. The fouling by alginate is attributed to the adsorption onto the membrane surface. At high ionic strength, the repulsive interaction

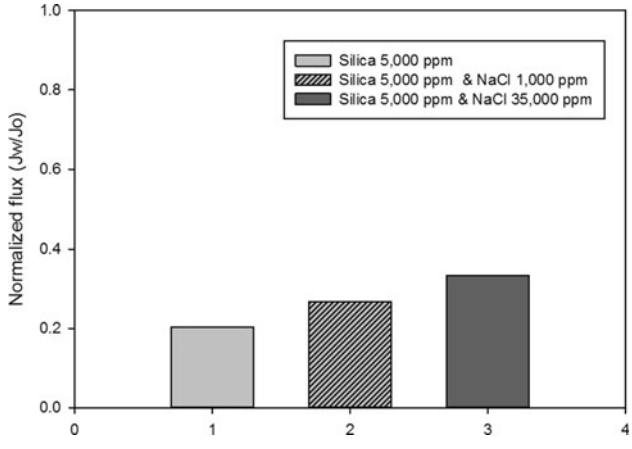

Fig. 5. Effect of NaCl concentration on flux decline by colloidal silica in FO system.

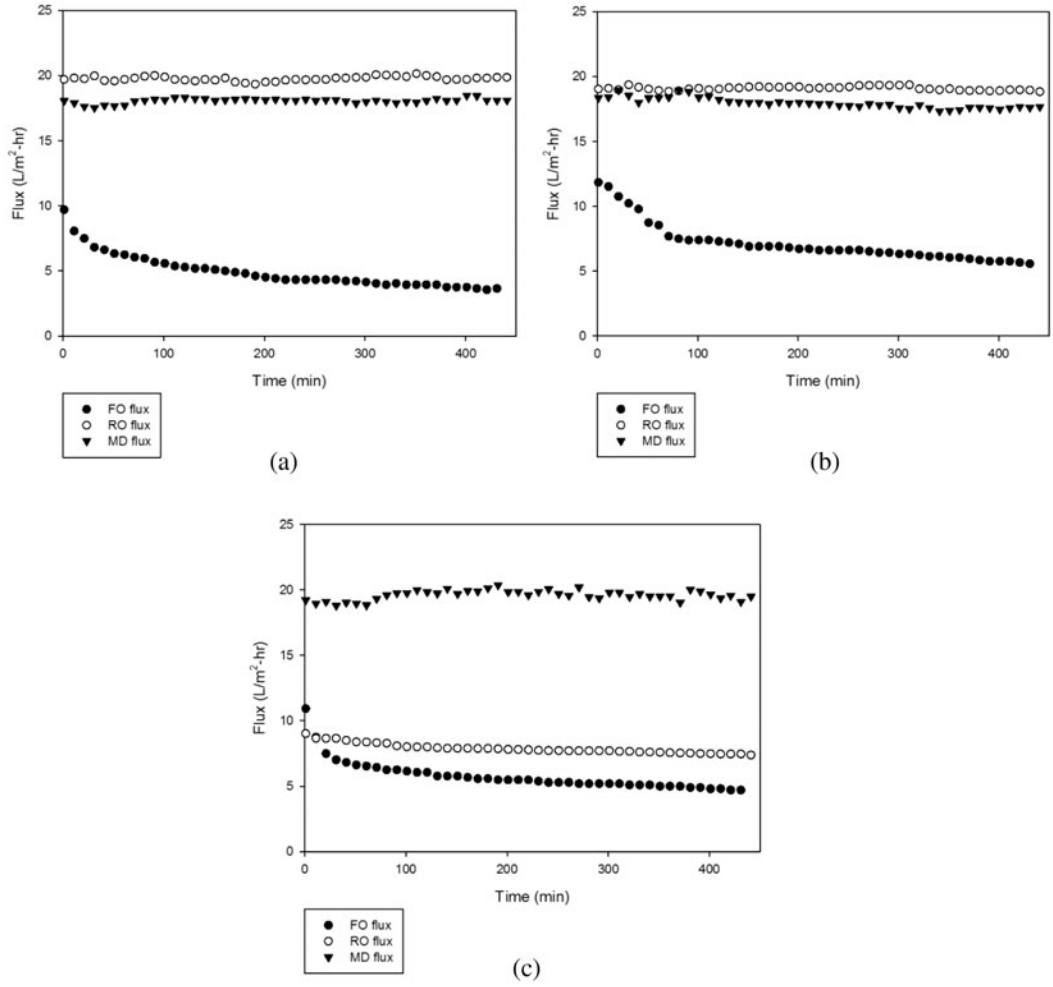

Fig. 6 Comparison of flux behaviors in FO, RO, and MD systems: (a) Feed water containing alginate of 500 mg/L, (b) Feed water containing alginate of 500 mg/L and NaCl of 1,000 mg/L, (c) Feed water containing alginate of 500 mg/L and NaCl of 35,000 mg/L, and (d) Normalized flux after the experiment.

between alginate and membrane surface may be reduced, leading to the adsorption and subsequent flux decline. As shown in Fig. 6, fouling by alginate occurs from the beginning, which is a typical behavior for adsorptive membrane fouling. The RSF in FO is the main reason why flux decline in FO system is the most serious. Fig. 7 shows the effect of NaCl concentration on the normalized flux after FO experiments using feed solution containing alginate. Unlike the case with silica (Fig. 5), the normalized flux values are similar regardless of the NaCl. When the feed solution contains only alginate, the adsorption can occur due to the RSF. As the NaCl concentration increases, the RSF may be reduced. Nevertheless, the solution near the membrane may have enough ion concentrations to induce the adsorption of alginate. In summary, the difference between silica and alginate is attributed to the difference in the fouling mechanisms.

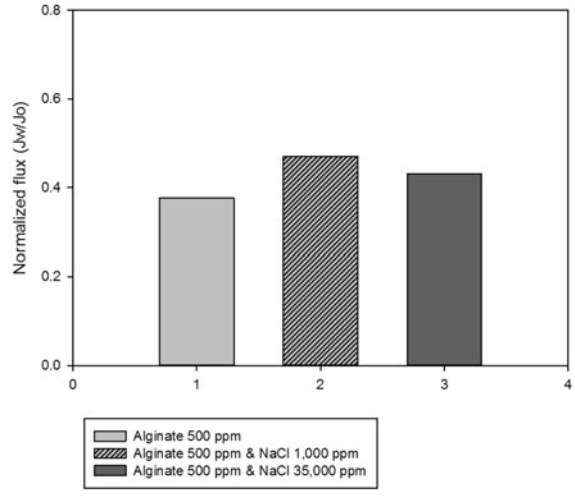

Fig. 7. Effect of NaCl concentration on flux decline by alginate in FO system.

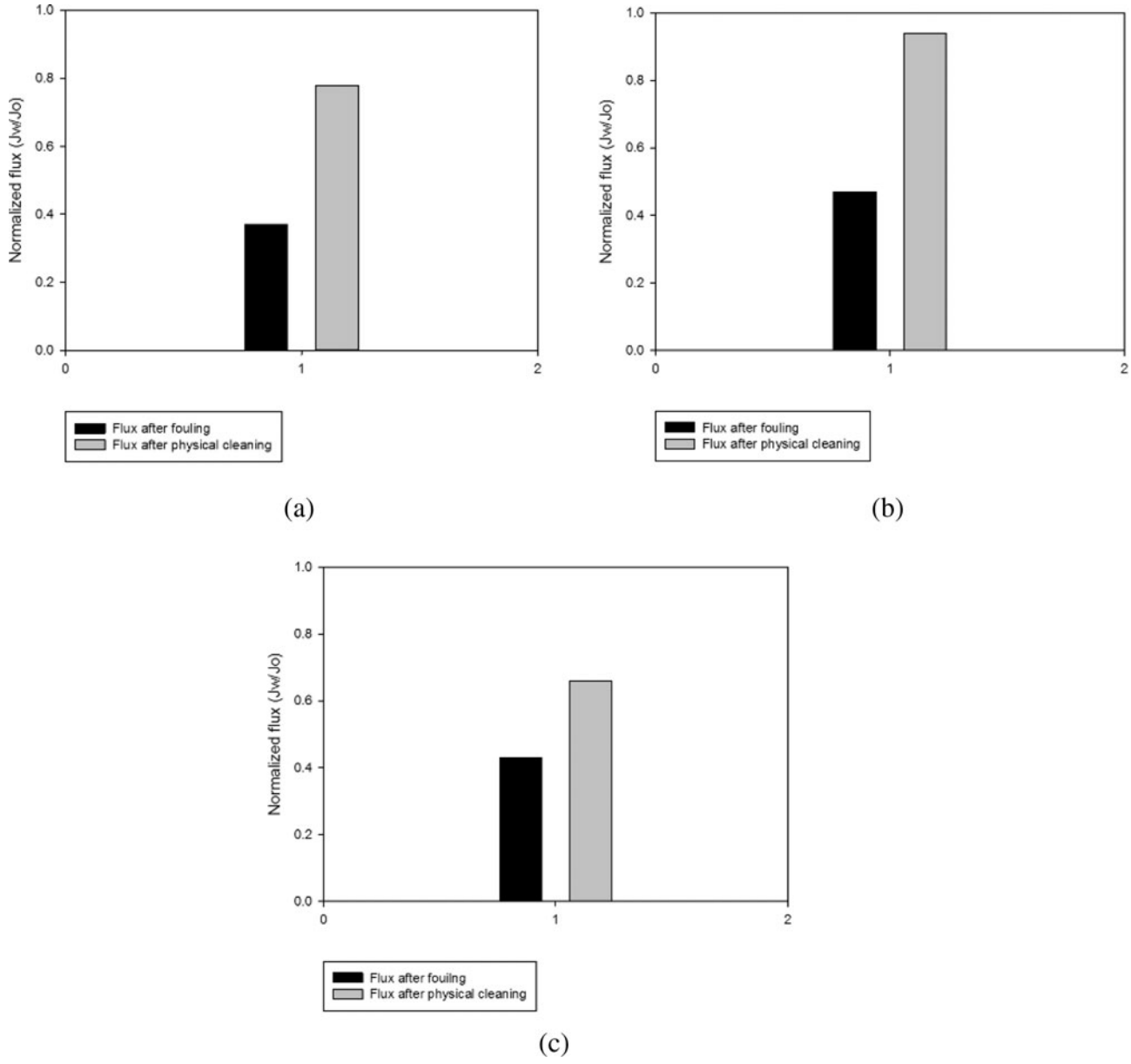

Fig. 8. Comparison of flux recovery for FO membranes fouled by colloidal silica: (a) Feed water containing colloidal silica of 5,000 mg/L, (b) Feed water containing colloidal silica of 5,000 mg/L and NaCl of 1,000 mg/L, and (c) Feed water containing colloidal silica of 5,000 mg/L and NaCl of 35,000 mg/L.

It should be noted that no fouling occurred in MD system by alginate. Since the MD membrane is hydrophobic, it was expected that adsorptive fouling also occurred. Nevertheless, it appears that the effect of organic adsorption on MD flux is not important. Since the vapor transports through the membrane in MD system, the adsorption of alginate may not affect it. An in-depth analysis will be required to further elucidate the sensitivity of MD flux to organic foulants.

3.3. Effect of physical cleaning on flux recovery

A physical cleaning method was applied to the fouled membranes. This was carried out using deionized water for 30 min at the recirculation flow rate of 1.0 L/min, which corresponds to 2 times higher than the flow rate in the fouling experiments. Fig. 8 compares the flux before and after physical cleaning for FO membranes fouled by colloidal silica. The recovery of flux by physical cleaning was high, which ranged

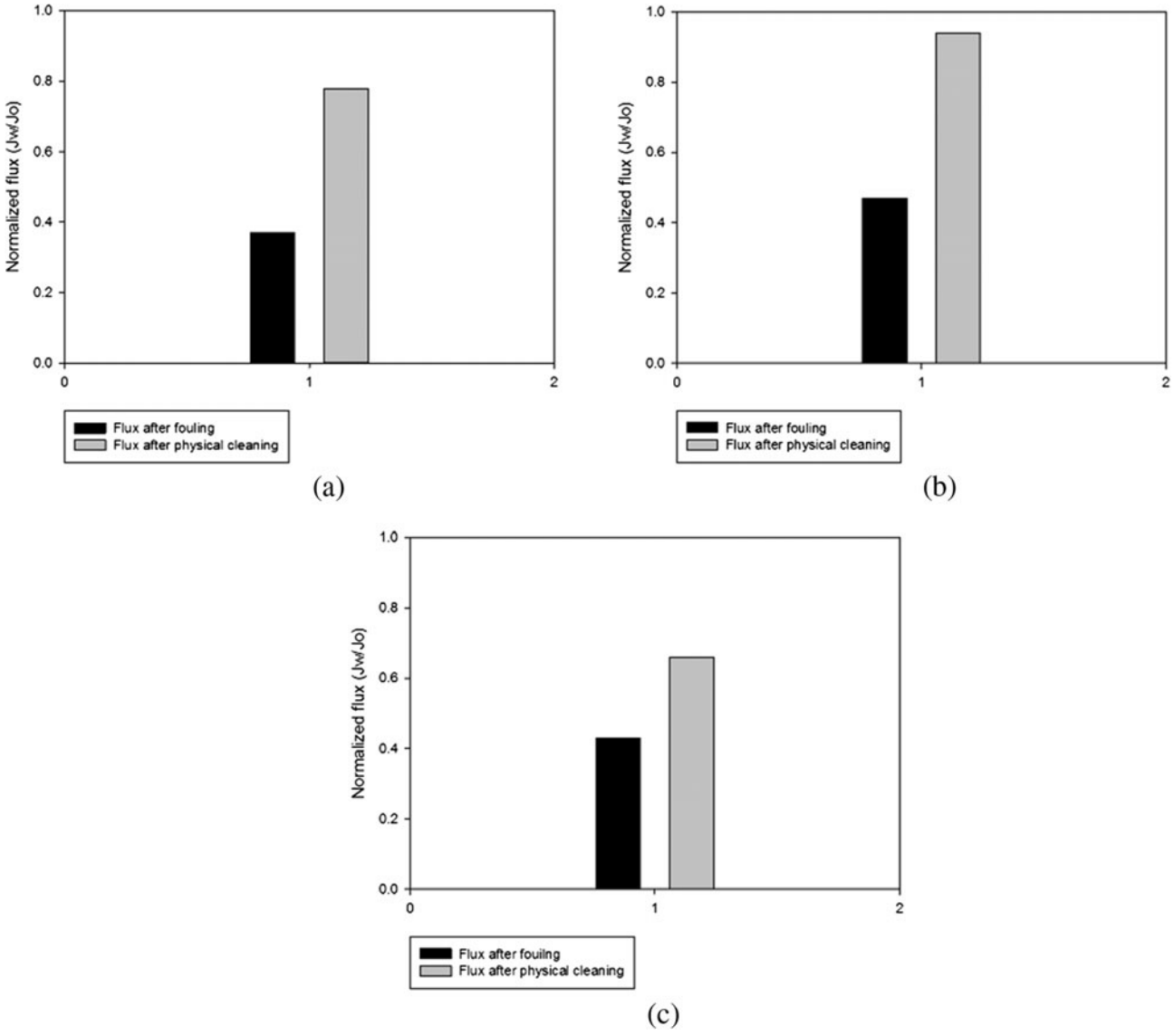

Fig. 9. Comparison of flux recovery for FO membranes fouled by alginate: (a) Feed water containing alginate of 500 mg/L, (b) Feed water containing alginate of 500 mg/L and NaCl of 1,000 mg/L and (c) Feed water containing alginate of 500 mg/L, and NaCl of 35,000 mg/L.

from 0.72 to 0.97. This suggests that the FO fouling by colloidal silica is reversible and the flux is easily recovered by simple physical flushing. Fig. 9 shows the flux before and after the physical cleaning for FO membranes fouled by alginate. The recovery of flux by the physical cleaning ranged from 0.65 to 0.95, which are similar to those in the case of colloidal silica fouling. It is likely that the fouling by alginate is also reversible.

The effects of the physical cleaning on the control of fouling by colloidal silica (5,000 mg/L) at high ionic strength (35,000 mg/L) are compared in FO, RO, and MD systems. As shown in Fig. 10(a) and (b), the normalized flux increased from 0.3 to 0.9 for the FO membrane and from 0.28 to 0.95 for the RO membrane. On the other hand, the flux recovery for the fouled MD membrane was not high: the normalized flux increased from 0.4 to 0.45 by applying the physical methods. This suggests that the fouling in MD membrane is less reversible than that in FO and RO membranes. This is attributed to the hydrophobic properties of MD membrane. It is likely that chemical cleaning methods should be applied to control fouling for MD membrane.

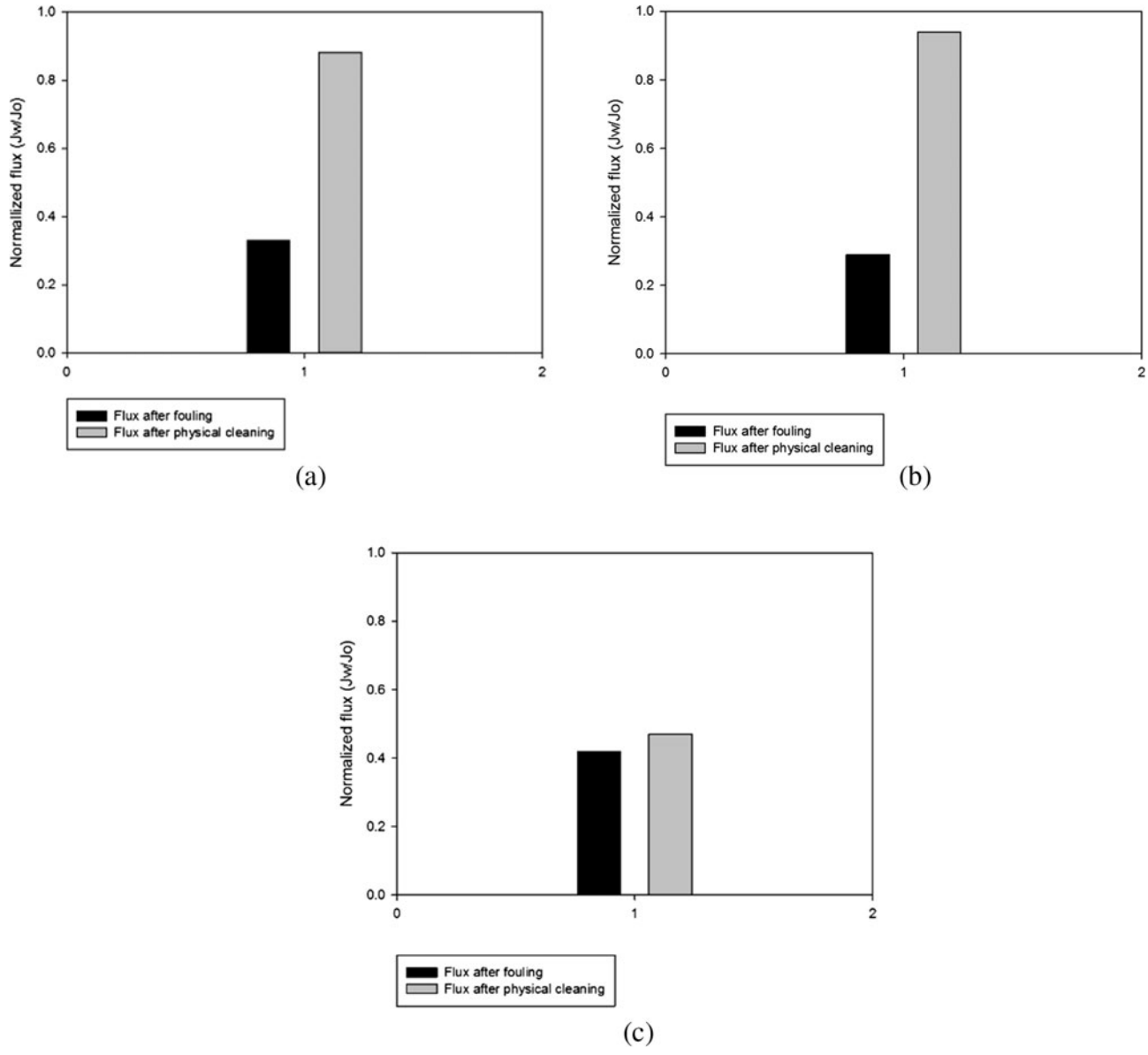

Fig. 10. Comparison of flux recovery for FO, RO, and MD membranes fouled by colloidal silica in NaCl solution of 35,000 mg/L: (a) FO membrane, (b) RO membrane, and (c) MD membrane.

4. Conclusion

In this study, the fouling and physical cleaning behaviors of FO, RO, and MD membranes were compared and the following conclusions were withdrawn:

(1) Colloidal silica resulted in membrane fouling for FO membrane while it did not cause flux decline in RO and MD membranes at low or moderate NaCl concentrations (<1,000 mg/L). At high NaCl concentration (35,000 mg/L), flux decline occurred in not only FO but also RO and MD membranes. Nevertheless, the fouling propensity was the lowest for MD membrane.

(2) Alginate also resulted in fouling for FO membrane. The flux decline immediately occurred from the beginning of the operation, suggesting that adsorption was the major fouling mechanism. Again, MD membrane showed the lowest fouling propensity.

(3) The fouling in FO membrane is attributed to the CEOP by RSF. Nevertheless, the flux was readily recovered by applying a simple physical cleaning method (i.e. flushing).
(4) Although MD membrane was less sensitive to fouling than FO and RO membranes, the recovery of flux by the physical cleaning was the lowest. This suggests that MD fouling is irreversible compared with FO and RO fouling.

Acknowledgment

This research was supported by a grant (12-TI-C01) from Advanced Water Management Research Program funded by Ministry of Land, Infrastructure and Transprot of Korean government.

References

[1] S.S. Shenvi, A.M. Isloor, A.F. Ismail, A review on RO membrane technology: Developments and challenges, Desalination 368 (2015) 10–26.
[2] V.G. Gude, Desalination and sustainability—An appraisal and current perspective, Water Res. 89 (2016) 87–106.
[3] A. Subramani, J.G. Jacangelo, Emerging desalination technologies for water treatment: A critical review, Water Res. 75 (2015) 164–187.
[4] N. Akther, A. Sodiq, A. Giwa, S. Daer, H.A. Arafat, S.W. Hasan, Recent advancements in forward osmosis desalination: A review, Chem. Eng. J. 281 (2015) 502–522.
[5] A. Deshmukh, N.Y. Yip, S. Lin, M. Elimelech, Desalination by forward osmosis: Identifying performance limiting parameters through module-scale modeling, J. Membr. Sci. 491 (2015) 159–167.
[6] N.M. Mazlan, D. Peshev, A.G. Livingston, Energy consumption for desalination—A comparison of forward osmosis with reverse osmosis, and the potential for perfect membranes, Desalination 377 (2016) 138–151.
[7] M. Qasim, N.A. Darwish, S. Sarp, N. Hilal, Water desalination by forward (direct) osmosis phenomenon: A comprehensive review, Desalination 374 (2015) 47–69.
[8] S. Liyanaarachchi, V. Jegatheesan, S. Muthukumaran, S. Gray, L. Shu, Mass balance for a novel RO/FO hybrid system in seawater desalination, J. Membr. Sci. 501 (2016) 199–208.
[9] L.D. Tijing, Y.C. Woo, J.-S. Choi, S. Lee, S.-H. Kim, H.K. Shon, Fouling and its control in membrane distillation—A review, J. Membr. Sci. 475 (2015) 215–244.
[10] D.M. Warsinger, J. Swaminathan, E. Guillen-Burrieza, H.A. Arafat, J.H. Lienhard V, Scaling and fouling in membrane distillation for desalination applications: A review, Desalination 356 (2015) 294–313.
[11] H.C. Duong, A.R. Chivas, B. Nelemans, M. Duke, S. Gray, T.Y. Cath, L.D. Nghiem, Treatment of RO brine from CSG produced water by spiral-wound air gap membrane distillation—A pilot study, Desalination 366 (2015) 121–129.
[12] H. Geng, J. Wang, C. Zhang, P. Li, H. Chang, High water recovery of RO brine using multi-stage air gap membrane distillation, Desalination 355 (2015) 178–185.
[13] N. Misdan, A.F. Ismail, N. Hilal, Recent advances in the development of (bio)fouling resistant thin film composite membranes for desalination, Desalination 380 (2016) 105–111.
[14] J.H. Jhaveri, Z.V.P. Murthy, A comprehensive review on anti-fouling nanocomposite membranes for pressure driven membrane separation processes, Desalination 379 (2016) 137–154.
[15] Q. She, R. Wang, A.G. Fane, C.Y. Tang, Membrane fouling in osmotically driven membrane processes: A review, J. Membr. Sci. 499 (2016) 201–233.
[16] Y. Ding, Y. Tian, J. Liu, N. Li, J. Zhang, W. Zuo, Z. Li, Investigation of microbial structure and composition involved in membrane fouling in the forward osmosis membrane bioreactor treating anaerobic bioreactor effluent, Chem. Eng. J. 286 (2016) 198–207.
[17] R. Honda, W. Rukapan, H. Komura, Y. Teraoka, M. Noguchi, E.M.V. Hoek, Effects of membrane orientation on fouling characteristics of forward osmosis membrane in concentration of microalgae culture, Bioresour. Technol. 197 (2015) 429–433.
[18] W. Xue, K. Yamamoto, T. Tobino, Membrane fouling and long-term performance of seawater-driven forward osmosis for enrichment of nutrients in treated municipal wastewater, J. Membr. Sci. 499 (2016) 555–562.
[19] M. Xie, J. Lee, L.D. Nghiem, M. Elimelech, Role of pressure in organic fouling in forward osmosis and reverse osmosis, J. Membr. Sci. 493 (2015) 748–754.
[20] R. Pamies, R.R. Schmidt, M.d.C.L. Martínez, J.G.d.l. Torre, The influence of mono and divalent cations on dilute and non-dilute aqueous solutions of sodium alginates, Carbohydr. Polym. 80(1) (2010) 248–253.
[21] T.H. Chong, F.S. Wong, A.G. Fane, Implications of critical flux and cake-enhanced osmotic pressure (CEOP) on colloidal fouling in reverse osmosis: Experimental observations, J. Membr. Sci. 314(1–2) (2008) 101–111.
[22] L.N. Sim, Y. Ye, V. Chen, A.G. Fane, Investigations of the coupled effect of cake-enhanced osmotic pressure and colloidal fouling in RO using crossflow sampler-modified fouling index ultrafiltration, Desalination 273 (1) (2011) 184–196.
[23] J.C. Eid, G.B. Andeen, Effects of acceleration on particulate fouling in reverse osmosis, Desalination 47(1–3) (1983) 191–199.
[24] W.C.L. Lay, J. Zhang, C. Tang, R. Wang, Y. Liu, A.G. Fane, Factors affecting flux performance of forward osmosis systems, J. Membr. Sci. 394–395 (2012) 151–168.
[25] C.Y. Tang, Q. She, W.C.L. Lay, R. Wang, A.G. Fane, Coupled effects of internal concentration polarization and fouling on flux behavior of forward osmosis membranes during humic acid filtration, J. Membr. Sci. 354(1–2) (2010) 123–133.