

Effect of silica fouling on RO membranes used for the desalination of Kuwait brackish water

Radhi Alazmi^{a,*}, Mubarak Alazemi^a, Adnan Alhathal Alanezi^a, Abdulsalam Alhazza^b

^aChemical Engineering Technology Department, College of Technological Studies, Public Authority for Applied Education and Training, P.O. Box: 42325, Shuwaikh 70654, Kuwait, emails: rh.alazmi@paaet.edu.kw (R. Alazmi), alazemmf@gmail.com (M. Alazemi), aa.alanezi@paaet.edu.kw (A. Alanezi)

^bWater Resources Division, Kuwait Institute for Scientific Research, P.O. Box: 24885, 13109 Safat, Kuwait, email: alhazza2@yahoo.com (A. Alhazza)

Received 7 July 2018; Accepted 13 June 2020

ABSTRACT

This study examined synthetic brackish water with inorganic foulants at concentrations similar to those in Kuwait brackish water. The concentration of silica was varied, and the effect of the change in the silica concentration on reverse osmosis (RO) membrane fouling was examined. Higher silica concentrations resulted in higher membrane fouling, and irreversible fouling increased dramatically when the silica concentration was increased beyond the solubility limit. Furthermore, the effect of the feed pH on the fouling of RO membranes by silica was also investigated. Maintaining a feed pH between 5 and 7 gave the lowest fouling. Finally, in general, an acidic cleaning solution led to better membrane recovery than a caustic cleaning solution. For two-step cleaning, the order of the cleaning steps was found to be highly important. Significant membrane recovery could be achieved with an initial caustic cleaning stage followed by an acidic cleaning stage.

Keywords: Brackish water; Membrane fouling; Silica RO fouling; Calcium RO fouling; RO membrane cleaning

1. Introduction

With the growing world population, the need for water for drinking, agriculture, and industry is rapidly increasing beyond that contained in the limited number of fresh water sources. The water shortage problem is strongly felt in arid regions of the world. Water scarcity is driving the quest to improve current water desalination technology and innovation. One of the most promising desalination technologies is reverse osmosis (RO). With rapid advancements in RO technology, the costs of constructing and operating RO desalination plants have been lowered. RO desalination solutions for sea and brackish water are gaining popularity worldwide [1]. One of the greatest advantages of RO desalination technology over conventional multi-flash distillation

technology is the reduced energy use [2]. For countries such as Kuwait that depend entirely on seawater desalination, it is sensible to implement RO desalination technology to produce fresh water. A large amount of oil can be saved by using RO technology, which consumes less energy than traditional multi-flash distillation technology [2].

In recent years, brackish water has been increasingly used for potable and agricultural purposes after treatment with RO desalination systems [3,4].

Nevertheless, the use of brackish water as a feed source for RO desalination systems is hindered by scale fouling with low-solubility salts, such as carbonates, sulfate minerals, and silica, that are present in brackish water [5–7].

The most pronounced disadvantage of RO technology is fouling of the membrane, which results in production losses

* Corresponding author.

and the need to change the membrane, which account for the largest operational costs in RO desalination plants.

Silica is considered to be one of the main RO membrane foulants and is very difficult to remove due to the irreversible nature of silica fouling [8,9]. Silica is naturally present in both amorphous and crystalline forms, which have different solubilities in water. Amorphous silica has a high solubility of 100–120 mg/L at 25°C and neutral pH, while crystalline silica has a low solubility of approximately 5–6 mg/L. Silica fouling of RO membranes can occur by one of two mechanisms: colloidal silica polymerizes in a bulk precipitate on the membrane surface, or individual monomeric silica polymerizes on the membrane surface [10]. Schulz et al. [11] reported severe membrane fouling and large reductions in hydraulic reversibility for artificial water containing model natural organic matter (humic acid, sodium alginate, and bovine serum albumin) and inorganic colloids (silicon dioxide and α -aluminum oxide) compared with the membrane fouling and hydraulic reversibility observed in experiments conducted with inorganic colloidal water only.

To overcome the fouling problem, carefully planned, and operated pretreatment and cleaning systems should be implemented. Because water quality is dependent on location, customized pretreatment, and cleaning systems should be adapted to the quality of the water being treated.

1.1. Chemical cleaning

Different materials can foul RO membranes, reduce the efficiency of RO membranes, or damage RO membranes. The main categories of fouling materials are inorganic foulants (i.e., particles and collides), organic foulants, and biofoulants.

The cleaning systems for each type of foulant are different [12,13]. Different fouling materials can be controlled with different chemicals; for example, carbonate-based foulants can be controlled by reducing the feed pH to 4–6, and sulfate-based foulants can be controlled with inorganic phosphate antiscaling [14].

Chemical cleaning solutions work differently for different organic and inorganic foulants. In general, acid-based cleaning solutions are used for inorganic foulants, and caustic solutions are used for organic foulants. Caustic and chelating agents have been very successful in recovering the

original flux of RO membranes fouled by organic materials [15]. Cleaning with ethylenediaminetetraacetic acid (EDTA) has been shown to be very effective in removing calcium-alginate scale from the surface of RO membranes, recovering 95% of the original flux [16].

In this study, the effects of changing the silica concentration and the feed pH on RO membrane fouling were investigated. Moreover, the performance effects of different cleaning solutions (chemicals and chemical concentrations) and cleaning order were studied.

2. Experimental procedure and methods

A Sterlitech Corporation CF042 crossflow filtration cell (Sterlitech Corporation, Kent, WA) was used in this study to perform all the filtration experiments (Fig. 1).

A new Dow FilmTec BW30 membrane (Dow FilmTec, Minneapolis, MN) was used in each experiment to ensure that irreversible fouling from previous experiments did not affect the membrane performance. The preparation procedure for each new membrane required soaking the membrane in deionized water overnight to remove any impurities from the manufacturing, packaging, and shipping processes. After soaking, the new membrane was installed in the filtration cell, and the system was run with deionized water for 3 h at constant pressure and temperature (20 bar and $22^\circ\text{C} \pm 1^\circ\text{C}$) to avoid any membrane compaction phenomena. Deionized water (EasyPure) was used to avoid altering the physical properties of the membrane. The deionized water was then replaced with 3 L of artificial brackish water (ABW) for each run. The ABW for the laboratory experiments was prepared by dissolving sodium metasilicate ($\text{Na}_2\text{SiO}_3 \cdot 9\text{H}_2\text{O}$) in 3 L of deionized water. The solution pH was then adjusted to approximately 7 using HCl and NaOH solutions. Calcium chloride, magnesium chloride, and sodium chloride were then added to simulate real brackish water. All chemicals used for preparing the ABW were of analytical grade (Table 1).

The system was maintained under a constant pressure of 20 bar by adjusting the membrane cell outlet needle valve. The temperature of the feed water was controlled using a custom-made ice-cooling outer bath with a temperature controller. The permeate from the RO filtration cell was collected in a graduated cylinder, which was placed on a digital balance. The data were continuously logged to a

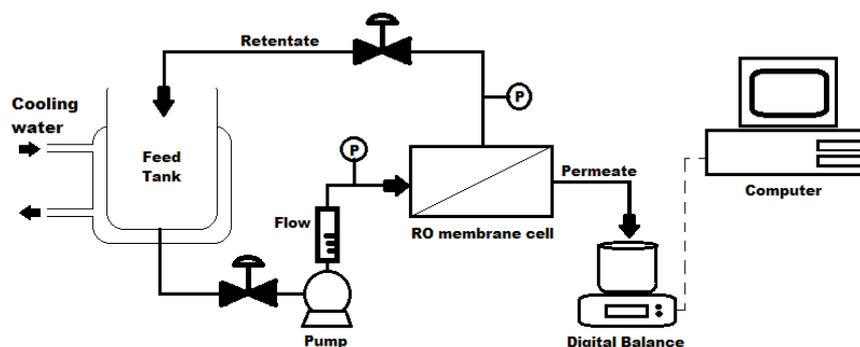


Fig. 1. CF042 crossflow filtration cell setup.

Table 1
Artificial brackish water composition

Chemical	Concentration (mg/L)
Sodium metasilicate	333.33–1,333.33*
Calcium chloride	1,000
Magnesium chloride	400
Sodium chloride	2,666.67

*Concentration of sodium metasilicate was different in each experiment.

computer using the data acquisition software ComDebug from Windmill Software Ltd.

Then, the effect of changing the ABW feed pH on the permeate flux was studied. The ABW flux experiments were prepared by dissolving sodium metasilicate ($\text{Na}_2\text{SiO}_3 \cdot 9\text{H}_2\text{O}$) in 3 L of deionized water. The solution pH was then adjusted to approximately 7 using HCl and NaOH solutions. Calcium chloride, magnesium chloride, and sodium chloride were then added to simulate real brackish water. The final pH was adjusted according to the experimental requirements using HCl and NaOH solutions.

The flux and membrane resistance were calculated using the following equations:

$$J = \frac{Q}{A_m} \quad (1)$$

where J is the flux (m/s), Q is the permeate flow (m^3/s), and A_m is the surface area of the membrane (m^2).

$$R = \frac{\Delta P}{\mu J} \quad (2)$$

where R is the resistance (1/m), ΔP is the transmembrane pressure (Pa), μ is the dynamic viscosity (Pa s), and J is the flux (m/s).

Scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDAX) analysis were performed on the surface of the used membranes using a JSM-6010LV InTouchScope, JEOL, Japan.

Table 2
Characteristics of the ABW

Parameter	Experiment # 1	Experiment # 2	Experiment # 3	Experiment # 4
Meta (mg/L)	334.47	669.63	1,001.50	1,335.47
CaCl_2 (mg/L)	1,000.80	1,000.93	1,000.87	1,001.93
MgCl_2 (mg/L)	401.50	400.63	402.67	400.50
NaCl (mg/L)	2,668.03	2,671.67	2,693.33	2,668.30
TDS (g/L)	3.17	3.78	3.84	4.10
NaCl (%)	12.4	14.8	15.0	16.0
EC (mS)	6.35	7.56	7.68	8.20
pH	7.00	7.09	6.93	7.10
SiO_2 (mg/L)	79.0	154.0	210.0	262.0

3. Results and discussion

3.1. Effect of silica concentration on the permeate flux

The effect of changing the silica dioxide concentration on membrane fouling was examined. Using the ABW described in Table 2, the concentration of silica dioxide was changed for each experiment to assess its effect on RO membrane fouling.

In general, the flux decreased as the concentration of SiO_2 increased (Fig. 2). The greatest reduction from the initial flux was observed for the experiment with an SiO_2 concentration of 262 mg/L. The high reduction was due to the deposition of precipitated silica on the membrane surface when the solubility limit was reached. This high silica concentration is typically reached at the end RO membrane elements of the brackish water desalination system operating on 70%–80% recovery of feed water. At this high silica concentration, the membrane permeate flux is greatly reduced, and the membrane resistance increases (Fig. 3). Moreover, the cost of operating the brackish water desalination system increases to a point where it would become economically unviable.

3.2. Effect of changing feed pH on membrane flux

Several studies have concluded that reducing the feed pH increases the permeate flux [17–19]. Lowering the pH of the feed ABW increases the solubility of salts. Increasing the solubility of salts reduces the chance of the salts reaching supersaturation and reduces the precipitation and crystallization of salts on the membrane surface.

In this study, the ABW feed pH was found to have a large effect on silica fouling. Maintaining a lower pH between 5 and 7 for the ABW feed reduced silica fouling and increased the permeate flux. Experiments conducted at a high pH (9–10) resulted in a poor permeate flux and a large increase in membrane resistance fouling (Fig. 4).

3.3. Efficiency of cleaning chemicals

3.3.1. One-stage cleaning

In this study, an acidic cleaning system and a caustic cleaning system, that is, a hydrochloric acid solution and a sodium hydroxide solution, respectively, were tested for

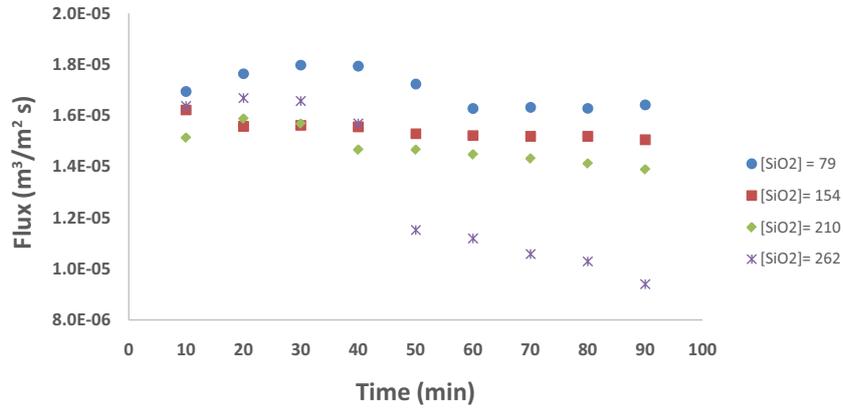


Fig. 2. Membrane flux vs. SiO₂ concentration in ABW feed experiments.

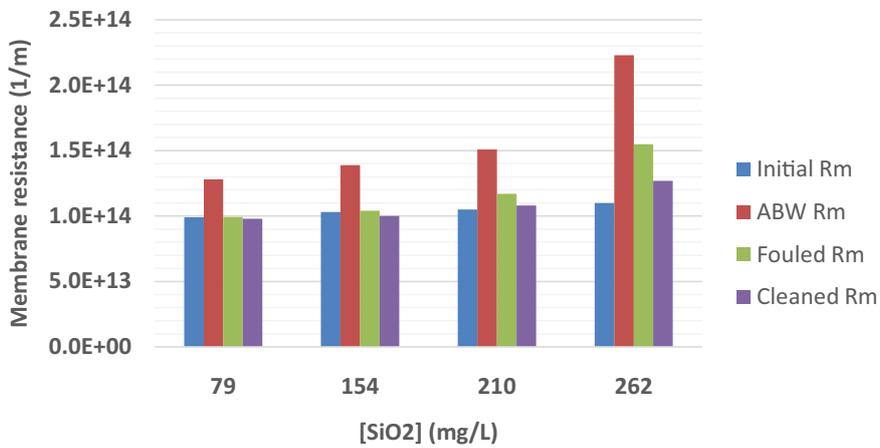


Fig. 3. Fouled membrane resistance vs. SiO₂ concentration in ABW feed experiments.

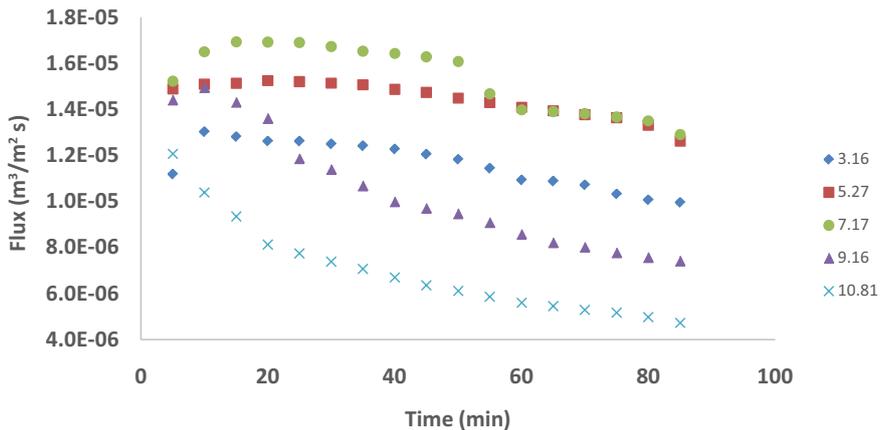


Fig. 4. Membrane flux at different ABW feed pH values.

cleaning silica-fouled RO membranes. Strong mineral acids such as HCl can solubilize deposits, dissolve them, and redispense foulants back into the bulk solution [20]. In caustic cleaning, the electrostatic repulsion and solubility of silicates are increased by NaOH because the silicate ionic strength is increased [21].

In general, the performance of the acidic cleaning system was better than that of the caustic cleaning system. The acidic cleaning system cleaned the fouled membrane at different fouling levels, achieving a high recovery of more than 80%. The caustic cleaning system gave worse results at high membrane fouling levels (Fig. 5). Visual inspection

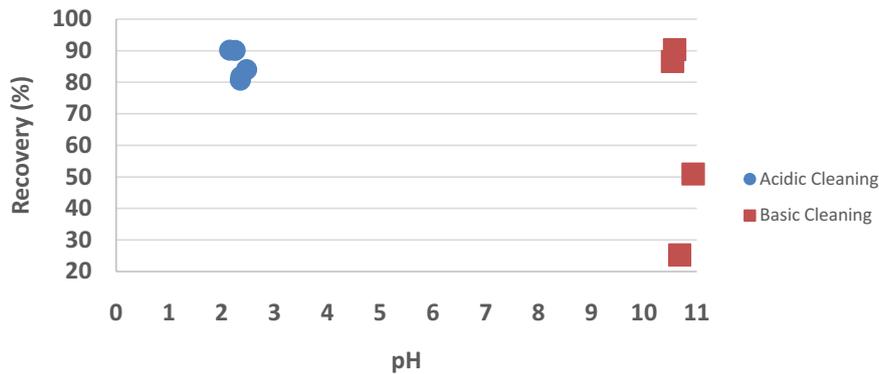


Fig. 5. Recovery efficiency of one-stage cleaning systems involving acidic or caustic cleaning solutions.

of the SEM images and foulant EDAX mass% concentration analysis show that the concentration of foulant attached on the RO membrane surface cleaned with HCl is lower than the concentration of foulant covering the RO membrane surface cleaned with NaOH (Fig. 6), with values of 20.73 ± 0.30 and 27.63 ± 0.65 (Table 3), respectively.

3.3.2. Two-stage cleaning

Two two-stage cleaning systems were investigated in this study – acidic cleaning for 30 min followed by caustic cleaning for 30 min and caustic cleaning for 30 min followed by acidic cleaning for 30 min – to determine if changing the cleaning order affects the efficiency of the two-stage cleaning process.

The best cleaning result was obtained with the two-stage cleaning system in which the first stage was a caustic solution and the second stage was an acidic solution (Table 4). The mass% concentration of foulants was lower after caustic-acidic cleaning than after acidic-caustic cleaning, with values of 0.48 ± 0.03 and 9.71 ± 0.38 , respectively (Table 3). The SEM images show that the concentration of foulant attached on the RO membrane surface cleaned with caustic-acidic system is lower than the concentration of foulant covering the RO membrane surface cleaned with acid-caustic system (Fig. 7).

Table 3
EDAX analysis of the RO membrane surface after cleaning

Cleaning system	Si concentration (%mass)	
Single cleaning	Caustic	27.63 ± 0.65
	Acidic	20.73 ± 0.30
Dual cleaning	Acidic-caustic	9.71 ± 0.38
	Caustic-acidic	0.48 ± 0.03

Increasing electrostatic repulsion and solubility of the silicates in the fouling layer coupled with the swelling effect of first-stage caustic cleaning of the membrane surface resulted in higher mass transfer for the second acidic cleaning stage, thus increasing the overall cleaning effect [22].

4. Conclusion

Silica is one of the most damaging foulants to RO membranes used for brackish water desalination and is very difficult to remove. The silica concentration found in real brackish water samples collected from the Wafra area south of Kuwait ranged from 34 to 45 mg/L. A brackish water desalination system operating at 70%–80% recovery

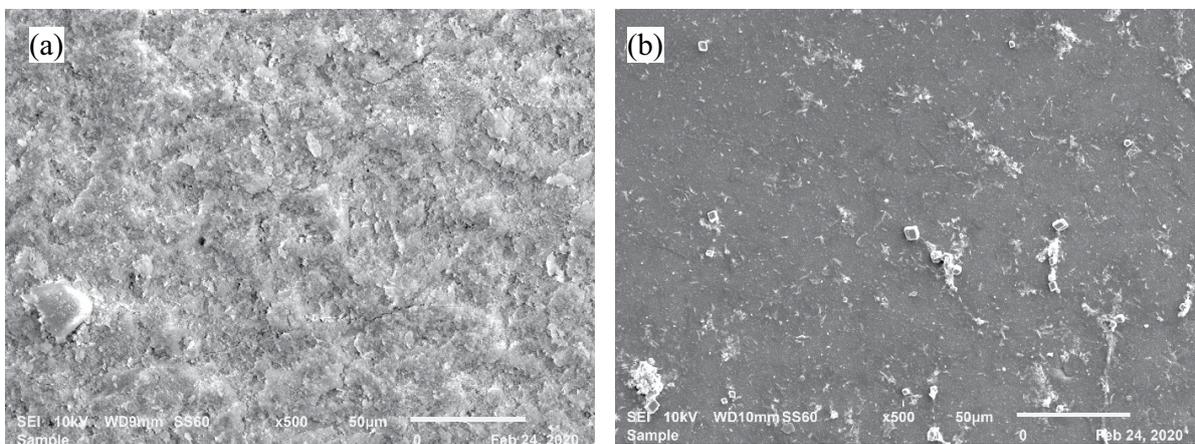


Fig. 6. SEM images of the RO membrane surface after (a) caustic and (b) acidic cleaning.

Table 4
Two-stage cleaning system recovery efficiency

First stage cleaning		Recovery (%)	Second stage cleaning		Recovery (%)
pH	Time (min)		pH	Time (min)	
10.50	30	85.60	2.11	30	101.33
10.88	30	93.22	2.35	30	100.00
10.54	30	86.61	2.25	30	99.10
2.47	30	84.06	10.55	30	96.67
2.15	30	90.18	10.63	30	93.52
2.35	30	80.65	10.56	30	91.74

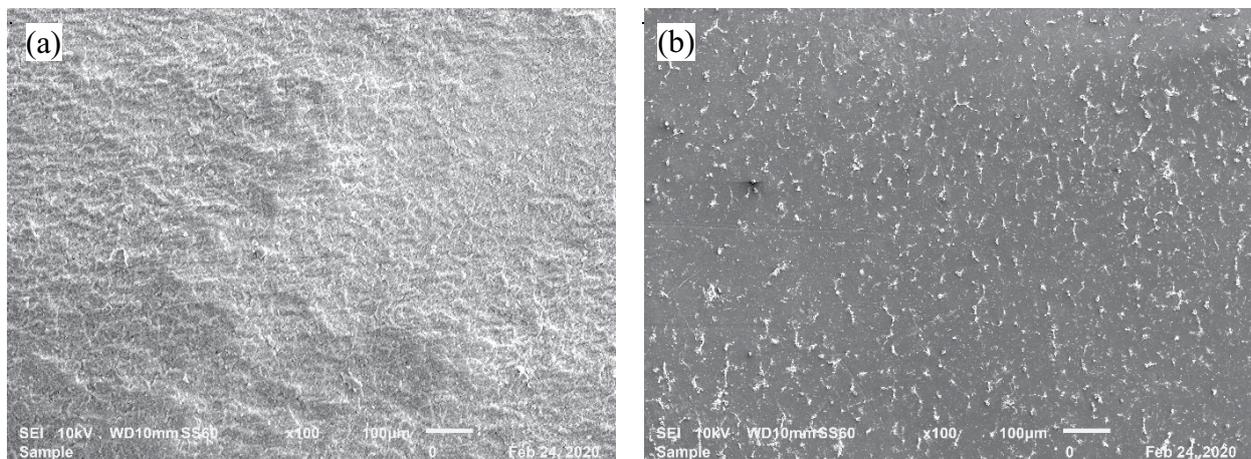


Fig. 7. SEM images of the RO membrane surface after (a) acidic-caustic and (b) caustic-acidic dual cleaning system.

would exceed the saturation level of silica (approximately 120 mg/L) at the last RO membrane element in the desalination system, causing irreversible damage to the RO membrane.

The brackish water feed pH was found to have a strong effect on silica fouling. Operating a brackish water desalination system with a feed pH between 7 and 5 would greatly improve the permeate flux and reduce silica fouling.

Finally, acidic cleaning was found to be more effective than caustic for cleaning the RO membrane and removing silica fouling. The best two-stage cleaning system to remove silica scale and recover the RO membrane was determined to be an initial caustic solution cleaning stage followed by an acidic solution cleaning stage. The first caustic cleaning stage increased the electrostatic repulsion and solubility of the silicates in the fouling layer, and the swelling effect of the caustic cleaning increased the mass transfer for the second acidic cleaning stage, resulting in higher overall cleaning performance.

Acknowledgments

This study was supported and funded by the Public Authority of Applied Education and Training, the College of Technological Studies, Research Project no: TS-14-06, Research Title: Effect of silica fouling on RO membranes used for the desalination of Kuwait brackish water.

References

- [1] S.H. Schneider, T.L. Root, M.D. Mastrandrea, *Encyclopedia of Climate and Weather*, Oxford University Press, New York, NY, 2011.
- [2] M.A. Darwish, N. Al-Najem, Cogeneration power desalting plants in Kuwait: a new trend with reverse osmosis desalters, *Desalination*, 128 (2000) 17–33.
- [3] 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.
- [4] B.C. McCool, A. Rahardianto, J. Faria, K. Kovac, D. Lara, Y. Cohen, Feasibility of reverse osmosis desalination of brackish agricultural drainage water in the San Joaquin Valley, *Desalination*, 261 (2010) 240–250.
- [5] M. Badruzzaman, A. Subramani, J. DeCarolis, W. Pearce, J.G. Jacangelo, Impacts of silica on the sustainable productivity of reverse osmosis membranes treating low-salinity brackish groundwater, *Desalination*, 279 (2011) 210–218.
- [6] G. Braun, W. Hater, C.Z. Kolk, C. Dupouiron, T. Harrer, T. Götz, Investigations of silica scaling on reverse osmosis membranes, *Desalination*, 250 (2010) 982–984.
- [7] E.M.V. Hoek, J. Allred, T. Knoell, B.-H. Jeong, Modeling the effects of fouling on full-scale reverse osmosis processes, *J. Membr. Sci.*, 314 (2008) 33–49.
- [8] T. Koo, Y.J. Lee, R. Sheikholeslami, Silica fouling and cleaning of reverse osmosis membranes, *Desalination*, 139 (2001) 43–56.
- [9] D. Lisitsin, D. Hasson, R. Semiat, Critical flux detection in a silica scaling RO system, *Desalination*, 186 (2005) 311–318.
- [10] P. Sahachaiyunta, T. Koo, R. Sheikholeslami, Effect of several inorganic species on silica fouling in RO membranes, *Desalination*, 144 (2002) 373–378.

- [11] M. Schulz, A. Soltani, X. Zheng, M. Ernst, Effect of inorganic colloidal water constituents on combined low-pressure membrane fouling with natural organic matter (NOM), *J. Membr. Sci.*, 507 (2016) 154–164.
- [12] E.G. Darton, M. Fazell, Presented at the 62nd Annual International Water Conference, Pittsburgh, 2001.
- [13] J. Xu, G. Ruan, X. Chu, Y. Yao, B. Su, C. Gao, A pilot study of UF pretreatment without any chemicals for SWRO desalination in China, *Desalination*, 207 (2007) 216–226.
- [14] R. Sheikholeslami, S. Tan, Effects of water quality on silica fouling of desalination plants, *Desalination*, 126 (1999) 267–280.
- [15] G.Z. Ramon, T.V. Nguyen, E.M.V. Hoek, Osmosis-assisted cleaning of organic-fouled seawater RO membranes, *Chem. Eng. J.*, 218 (2013) 173–182.
- [16] W.S. Ang, A. Tiraferri, K.L. Chen, M. Elimelech, Fouling and cleaning of RO membranes fouled by mixtures of organic foulants simulating wastewater effluent, *J. Membr. Sci.*, 376 (2011) 196–206.
- [17] I.H. Huisman, P. Prádanos, A. Hernández, The effect of protein–protein and protein–membrane interactions on membrane fouling in ultrafiltration, *J. Membr. Sci.*, 179 (2000) 79–90.
- [18] A. Maartens, P. Swart, E.P. Jacobs, Feed-water pretreatment: methods to reduce membrane fouling by natural organic matter, *J. Membr. Sci.*, 163 (1999) 51–62.
- [19] C.H. Koo, A.W. Mohammad, F. Suja', M.Z. Meor Talib, Review of the effect of selected physicochemical factors on membrane fouling propensity based on fouling indices, *Desalination*, 287 (2012) 167–177.
- [20] S.S. Madaeni, Y. Mansourpanah, Chemical cleaning of reverse osmosis membranes fouled by whey, *Desalination*, 161 (2004) 13–24.
- [21] K.J. Kim, P. Sun, V. Chen, D.E. Wiley, A.G. Fane, The cleaning of ultrafiltration membranes fouled by protein, *J. Membr. Sci.*, 80 (1993) 241–249.
- [22] N. Porcelli, S. Judd, Chemical cleaning of potable water membranes: a review, *Sep. Purif. Technol.*, 71 (2010) 137–143.