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A comparative study on fouling behaviors of hollow fiber membranes in pressure-driven and temperature-driven operations

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ABSTRACT

Hydrophobic hollow fiber membranes have been used for pressure-driven separation processes such as microfiltration (MF) and ultrafiltration. Recently, they are also applied for membrane distillation (MD) that uses thermal energy as its driving force. In this study, the fouling characteristics of the MD process were compared with those of the MF process using the same hollow fibre membranes. Colloidal silica and alginate were used as model foulants for both cases. Submerged MF and direct contact MD were implemented using laboratory-scale equipments. Results showed that MD fouling due to the colloidal silica or alginate could not be correlated with silt density index or modified fouling index. Moreover, the fouling behaviors and foulant layers were also different between MF and MD although same membranes and feedwaters were used. This is attributed to the effect of applied hydraulic pressure that only exists in MF process.

Keywords: Hollow fiber; Membrane distillation; Microfiltration; Membrane fouling; Fouling index

1. Introduction

The shortage of available water is a serious problem in this century. Although the total freshwater supply does not change, water demands are rapidly increasing due to population growth and industrialization [1]. Accordingly, water supply becomes inadequate to match the increased water demands. Moreover, climate change also reduces the amount of available water resource by decreasing river flow, and shrinking lakes and reservoirs. Water pollution is also one of the reasons to reduce available freshwater [1]. Accordingly, seawater desalination becomes one of the leading technologies to solve the problems related to water shortage [2–4].

While the reverse osmosis (RO) membrane is now widely adopted for many desalting applications including seawater desalination, membrane distillation (MD) is one of the emerging desalination technologies for the production of freshwater [5,6]. MD is a thermally-driven separation process, in which only vapor molecules are able to pass through a porous hydrophobic membrane [7–12]. MD has many advantages over conventional desalination technologies: first, the operating temperature for MD is lower than those for other evaporation processes such as multistage flash and multieffect distillation. Second, the applied pressure for MD is lower than that for RO, allowing the off-grid operation. Using MD, it is also possible to achieve almost complete rejection of non-volatile contaminants including salts and dissolved organic matters [13,14]. In addition, MD can be used for water recovery from high-salinity solutions like RO brine [6,11,12,15].

However, like all other membrane processes, a major inefficiency of MD is fouling, which causes a decline in membrane permeability due to the accumulation of deposits on the membrane surface and inside the membrane pores [5,16,17]. In RO and nano filtration (NF) processes, the silt density index (SDI) and the modified fouling index (MFI) are the existing methods to measure the particulate fouling potential [4,18–20]. However in case of MD, the issue on fouling is still not well understood, due to differences in membrane structure and operational conditions. As fouling is important issue

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that should be addressed to enhance the efficiency of MD process, there is a need to understand its formation mechanism, and the different parameters that affect its propensity and possible mitigation or cleaning strategies [5].

Accordingly, the primary objective of this study was to understand fouling characteristics in MD processes. Colloidal silica and alginate were used as model foulants. Because SDI and MFI are the standardized parameters and widely used in engineering practices [19,21], the concentrations for fouling experiments are determined based on SDI and MFI values of each foulants. Prior to the experiments, the relationships between foulants concentration and SDI/MFI were obtained. In this study, direct contact membrane distillation (DCMD) system was employed same hollow fiber hydrophobic MD membranes were used in both MD and microfiltration (MF) operations for the treatment of same feedwater. After fouling experiments membrane surfaces were observed by scanning electron microscope (SEM).

2. Materials and methods

2.1. Silica and alginate

Commercially available silica (SiO_2) particles, LUDOX TMA-30 colloidal silica (Sigma-Aldrich, South Korea) were used for fouling index experiments. The average particle size of the silica is 22 nm [22] and maximum surface area is 140 m^2/g. Because concentration of this colloidal silica is 34 wt%, stock solution (10 mg/L) was prepared by dilution with deionized (DI) water.

Moreover, medium viscosity alginic acid sodium salt from brown algae (Sigma-Aldrich, South Korea) was used as an organic foulant. The form of alginate is powder and the molecular weight is $3.50 \pm 0.04 \times 10^5$ g/mol [23]. Alginate stock solution (10 mg/L) was also prepared by dilution with DI water.

2.2. Laboratory submerged membrane filtration system

A photograph of laboratory submerged membrane filtration system is shown in Fig. 1. Feedwater is filled in 1 L volume acrylic tank and recirculated by multichannel pump (EW-07551-00, Cole-Parmer, USA). When feedwater is sucked by pump, transmembrane pressure (TMP) is presented in the digital pressure gage (ISE40A-01-R, SMC, Japan) and recorded in every second in the desktop. The system consisted of 15 filtration tanks, allowing test can be carried out with various feed conditions simultaneously. Magnetic stirrer at bottom of the tanks makes feedwater dispersed consistently. Pump was operated by constant flux mode. Pump was operated in 40, 80 and 120 LMH to find specific operating condition which can identify distinct TMP increase. Experiments were carried out for 3 h for every fluxes and foulants conditions.

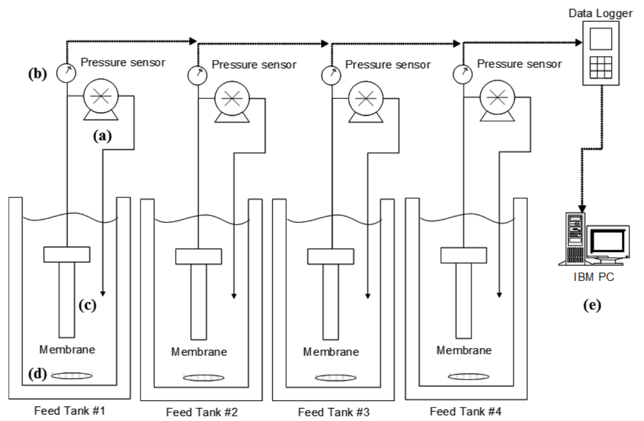


Fig. 1. Experimental setup for submerged MF test: (a) multichannel pump, (b) digital pressure gage, (c) hollow fiber membrane, (d) magnetic stirrer and (e) desktop for data logging.

2.3. SDI and MFI test

The SDI test was carried out by passing feedwater through 0.45 μm filter with constant pressure (2 bar). In the SDI test, SDI_{15min} was adopted. The SDI values were calculated from the equation:

$$\mathrm{SDI} = \frac{1 - \frac{t_f}{t_i}}{\Delta t} \times 100 \quad (1)$$

where t_i is the time to collect initial 500 mL of sample, t_f the time to collect final 500 mL of sample (usually 15 min) [24], Δt the total running time for the test [18].

The MFI test was also carried out at the same time by recording flux decline rate until the end of operation. The definition of MFI is as follows:

$$\mathrm{MFI} = \frac{\alpha C_s \eta}{2 \Delta P A^2} \quad (2)$$

where α is the specific cake resistance, C_s the bulk concentration of suspended solids, ΔP the transmembrane pressure and A the area of the membrane used for the test [18]. Schematic diagram of lab scale SDI/MFI device is shown in Fig. 2.

2.4. Laboratory MD system

The driving force of DCMD is a temperature-induced vapor pressure difference caused by having a hot feed and a cold permeate [13,25]. Laboratory DCMD system and direction of water flow are shown in Fig. 3.

Heated feed and cold permeate water flow by countercurrent flow in the membrane module. Temperature of feed and

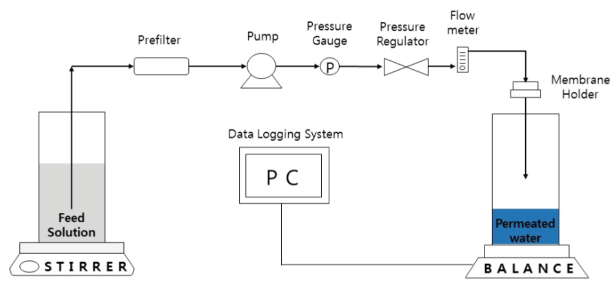


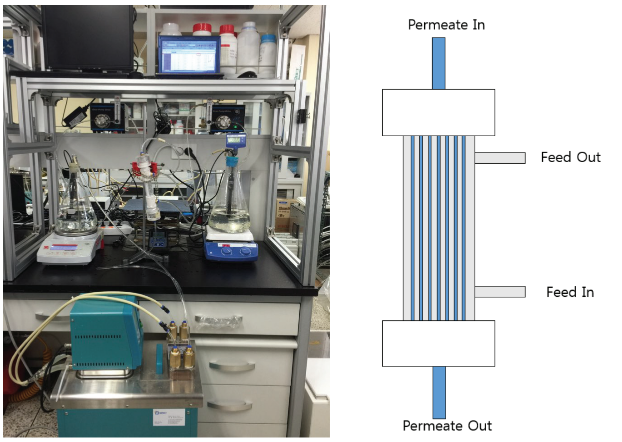
Fig. 2. Schematic diagram of lab scale SDI/MFI setup.

permeate water was 60°C and 20°C, respectively, with 40°C temperature difference. After passing through the module, permeate was in and out the chiller maintained 20°C by water bath. Pump (Micro gear pump) was operated in constant flow rate – 0.4 L/min for feed and 0.6 L/min for permeate.

2.5. MD membrane

In this study, hollow fiber polyvinylidene fluoride (PVDF) hydrophobic membranes (Econity, South Korea) were used. The nominal pore size of the membrane was 0.22 μm. The inner and outer diameters were 0.7 and 1.3 mm, respectively. Ten hollow fiber membranes for 10 cm length were prepared and effective membrane area for the tests was 0.00314 cm^2. Average flux in DI water test for the membrane modules was about 10–11 LMH.

Before submerged membrane tests, the membranes had modifying process to change hydrophobic property into hydrophilic property by soaking in alcohol.



(a) (b)

Fig. 3. Laboratory DCMD system setup and flow of water in module: (a) experimental setup and (b) hollow fiber MD module.

3. Results and discussion

3.1. SDI and MFI tests

SDI and MFI tests were conducted using colloidal silica and alginate. Feed concentrations used in the experiments range 30–240 ppm for silica and 10–250 ppm for alginate. Table 1 shows the results of experiments including t_i, t_f and SDI values and fouling index (both SDI and MFI) graph is shown in Fig. 4.

The results show that higher fouling index values are observed when feed solutions have higher concentration. As shown in Fig. 4, fouling indices of silica increase nearly in a linear fashion and that of alginate increase in a log form. Using linear regression method, a correlation between foulant concentration and SDI/MFI were obtained with $R^2 > 97\%$. Based on the results, feed concentrations for MD tests were determined, which are summarized in Table 2.

3.2. Submerged MF test

Submerged MF experiments using the hollow fiber MD membranes were carried out to compare fouling propensity with that of DCMD. The experiments flux conditions were 40, 80 and 120 LMH and each test was done for 3 h. The results of TMP variations are shown in Fig. 5.

As shown in Fig. 5, silica has no effect on fouling in all fluxes and concentration conditions. On the other hand, with an increase in alginate concentrations in over 120 LMH range, TMP becomes higher and the gradient of TMP is also bigger. Even in a very low concentration, alginate causes membrane fouling. From the results, only alginate can be matched with SDI/MFI in submerged membrane system.

3.3. DCMD test

The results for DCMD experiments using silica and alginate as model foulants are shown in Fig. 6. In each condition, DCMD experiment was carried out for 50 h. After the experiment, the surfaces of MD membranes were examined, which are shown in Fig. 7. These results were compared with the SDI values and MF results.

Table 1
SDI values according to silica and alginate concentrations

Silica				Alginate			
Concentration (ppm)	t_i	t_f	SDI_{15}	Concentration (ppm)	t_i	t_f	SDI_{15}
30	74	85	0.86	10	124	193	2.38
60	71	98	1.84	20	110	210	3.17
90	74	116	2.41	40	97	284	4.39
120	74	160	4.38	60	88	309	4.77
150	73	213	3.58	80	111	428	4.94
180	71	354	5.33	100	85	507	5.55
210	77	481	5.6	120	79	536	5.68
240	69	945	6.18	150	97	703	5.75
				200	134	1,105	5.86
				250	129	1,445	6.07

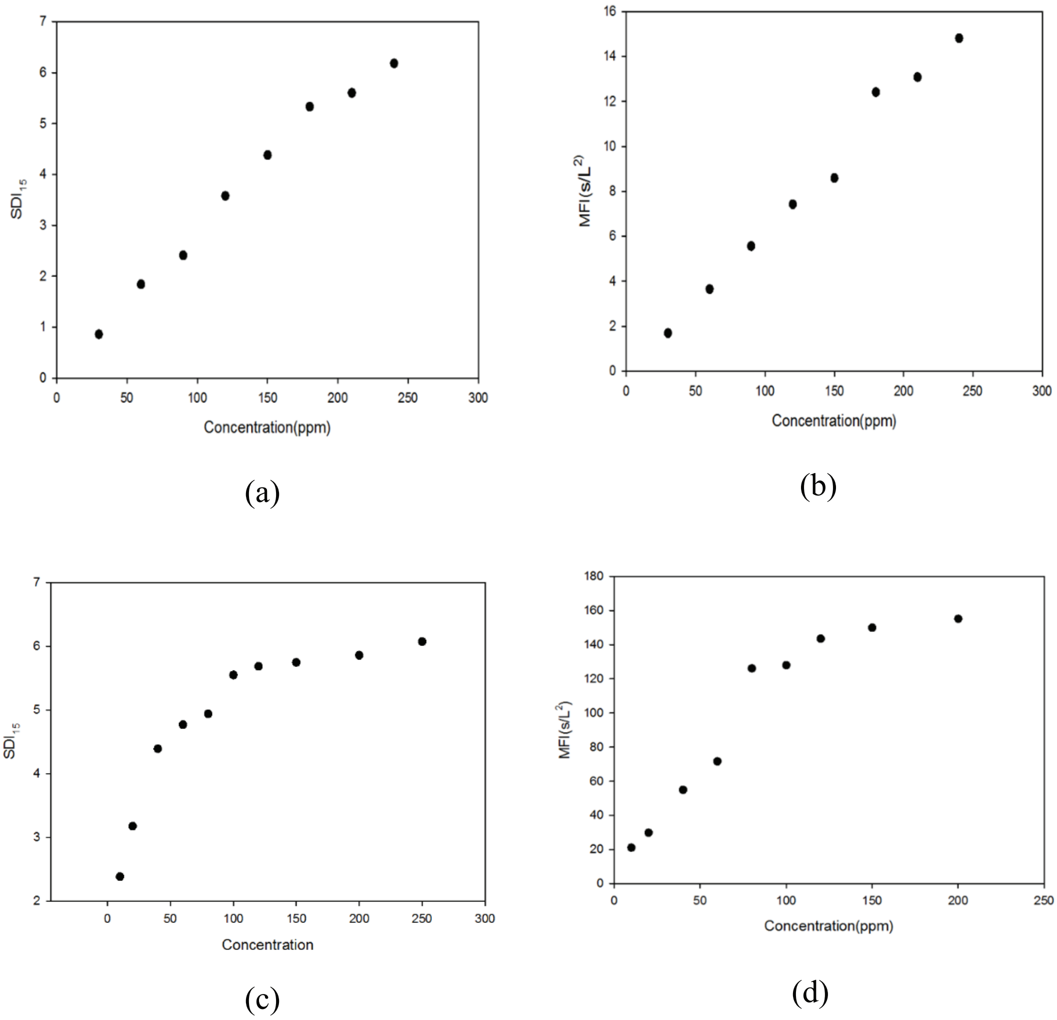


Fig. 4. Fouling index (SDI and MFI) graphs by feed concentrations: (a) SDI of silica, (b) MFI of silica, (c) SDI of alginate and (d) MFI of alginate.

Table 2
Determined feed concentration conditions for MD experiments

Silica		Alginate	
Concentration for experiments (ppm)	SDI	Concentration for experiments (ppm)	SDI
50	1.46	1.8	0.49
120	3.60	6.4	2.00
250	6.52	23	3.51
		85	5.06
		285	6.50

Figs. 6(a)–(c) show the dependence of MD flux on time at different silica concentrations. It is evident from the graphs that fouling was negligible regardless of silica concentration. It should be noted that SDI increases from 1.46 to 6.52 with increasing silica concentration from 50 to 250 mg/L. This implies that SDI cannot predict the fouling potential for MD by colloidal silica.

On the other hand, the results from submerged MF tests showed no fouling under these conditions, which can be correlated with the results of MD tests. As shown in Figs. 5(a)–(c), MF flux did not decrease in all cases. This is attributed to the surface property of the membrane and foulants. Since the membranes used for MF and MD have low surface energy, the interaction between the membrane and foulant may be small. Since SDI tests were done under dead-end filtration mode, the interaction between the membrane and foulants are not important. On the other hand, the MF and MD tests were done in semi-crossflow or crossflow conditions, the effect of interaction may become important.

Figs. 6(d)–(h) show the changes in MD flux with time at different alginate concentrations. Although it is not clearly shown, the flux slightly decreased at the end of the experiment. Nevertheless, the fouling propensity was not significant even after 50 h operation. Again, the SDI value, which is proportional to alginate concentration, cannot be correlated with this fouling tendency.

For alginate, the results in the MF operation were also compared with those in the MD operation. Unlike silica,

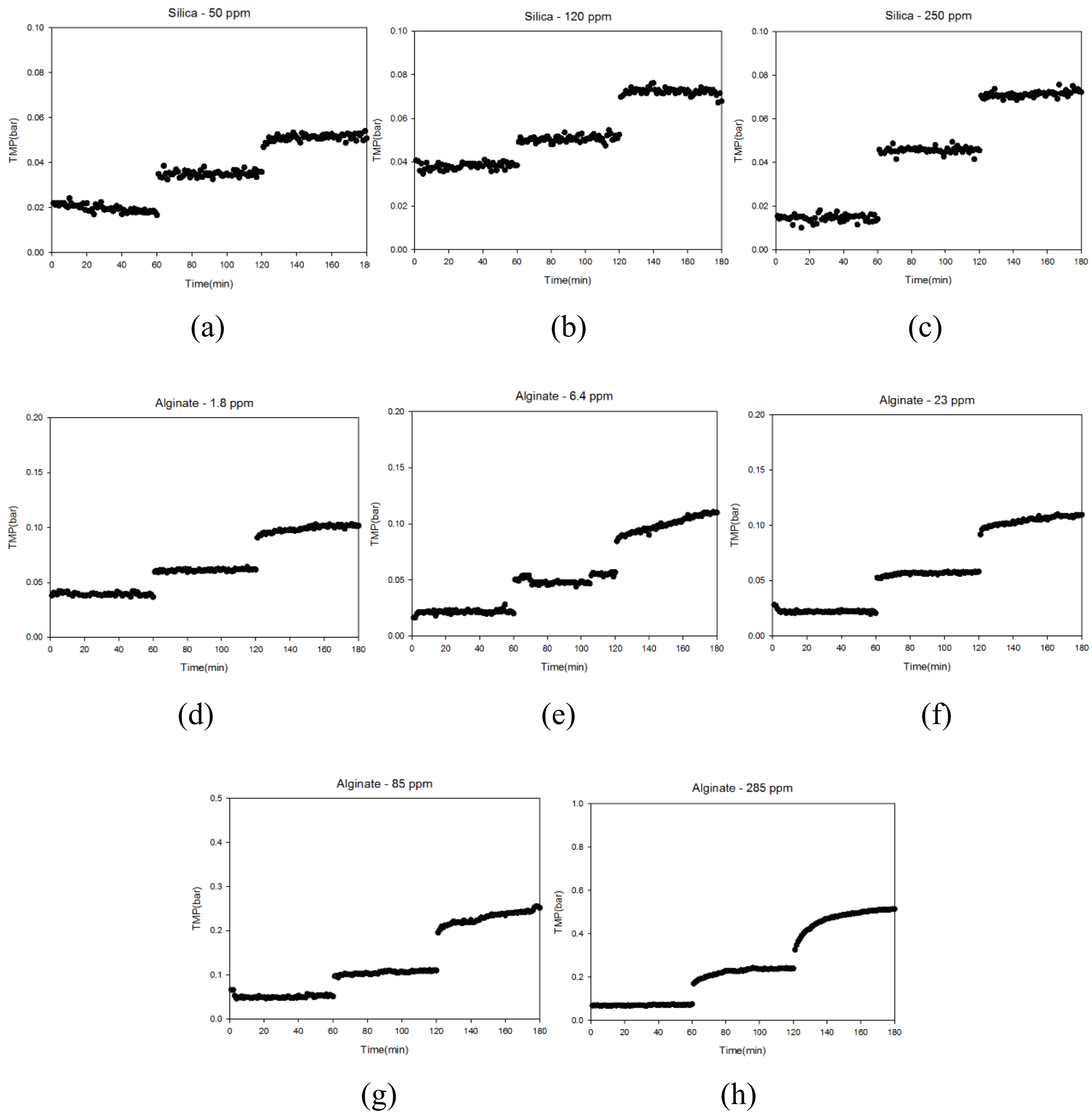


Fig. 5. Variation of TMP according to types of feed and feed concentrations: (a) silica 50 ppm, (b) silica 120 ppm, (c) silica 250 ppm, (d) alginate 1.8 ppm, (e) alginate 6.4 ppm, (f) alginate 23 ppm, (g) alginate 85 ppm, (h) alginate 285 ppm.

the TMP of MF increases with an increase in alginate concentration and operation flux. This implies that alginate can cause fouling in this membrane. Nevertheless, quantitative interpretation of fouling potential in MD by this MF method was not possible. This is probably because of the different mechanisms of MD and MF: MD is a thermally-driven membrane process without any pressure effect. On the contrary, MF is a pressure-driven process, in which pressure may affect fouling rate. Since alginate can form gel layer on the membrane surface during MF, an increase in pressure will result in gel layer compaction and accelerate fouling. Accordingly, it is likely that the fouling in MF by alginate is faster than that in MD.

Fig. 7 shows the SEM images for the MD membranes after DCMD fouling tests using silica or alginate. As silica concentration increases, the coverage of cake layer on the MD membrane increases (Figs. 7(b)–(d)). As mentioned above, the silica did not cause flux decline in MD even at its high concentration. Accordingly, it is evident that the cake layer formation by silica and MD fouling is not related. This is also the reason why SDI fails to predict fouling potential in MD.

On the other hand, it was difficult to identify alginate on the membrane surface after the DCMD experiment. This is attributed to the fact that alginate gel layer was not formed on the membrane surface in MD operation. After SDI test or MF test, the alginate generally form gel layers to decrease permeability of the filter or membrane. However, such phenomena cannot be found after MD test. It is also suggested that alginate can be easily washed away from membrane surfaces by shear force generated when feed solutions pass on the membrane surface. This strongly suggests that there is a clear difference in fouling mechanism between pressure-driven and thermally-driven membrane processes.

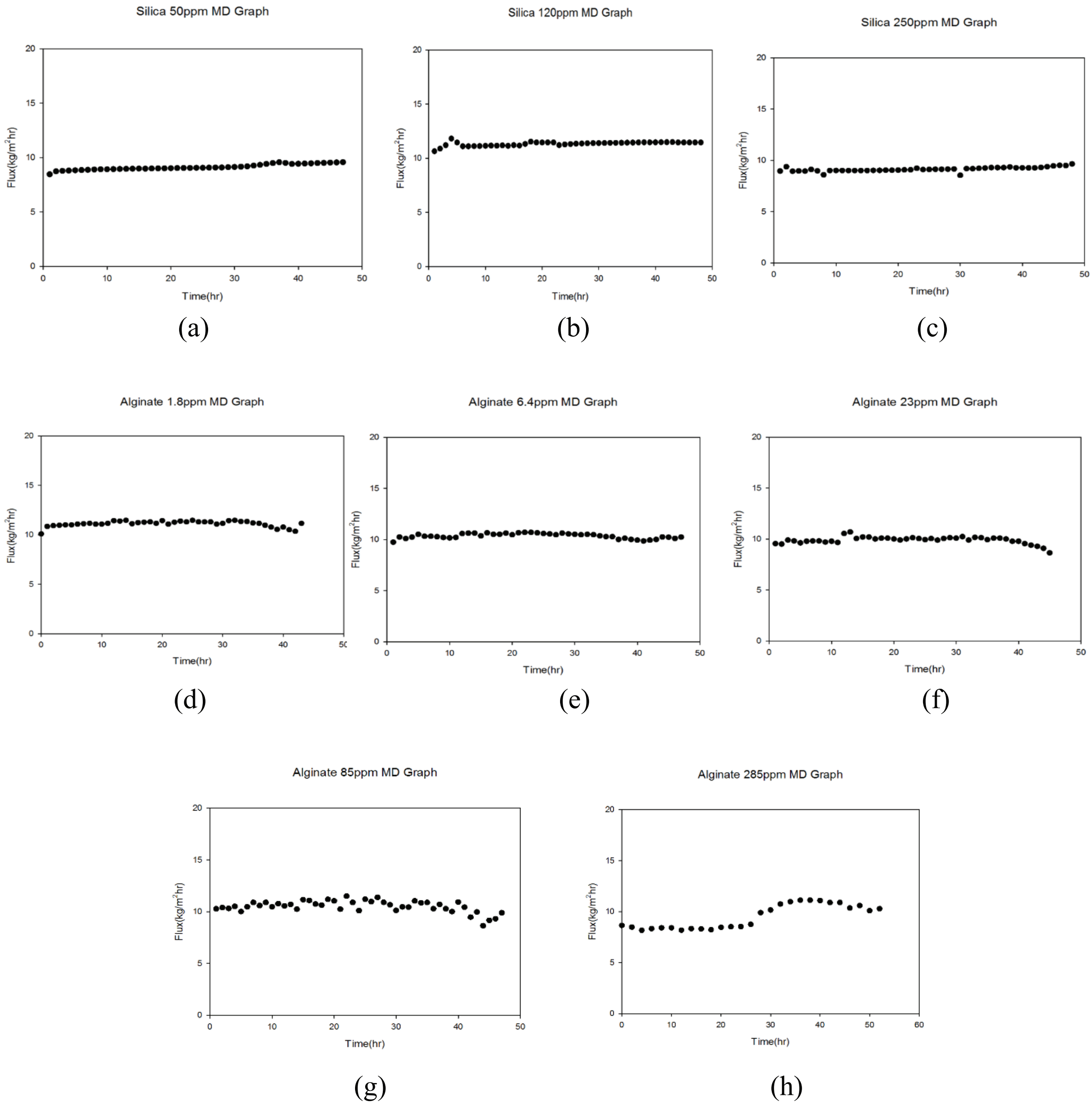


Fig. 6. DCMD fluxes according to types of feed and feed concentrations: (a) silica 50 ppm, (b) silica 120 ppm, (c) silica 250 ppm, (d) alginate 1.8 ppm, (e) alginate 6.4 ppm, (f) alginate 23 ppm, (g) alginate 85 ppm, (h) alginate 285 ppm.

4. Conclusion

In this study, fouling behaviors of hollow fiber membranes in MF (pressure-driven operation) and MD (temperature-driven operation) were compared. The following conclusions were drawn:

- In DCMD experiments, no fouling was observed by silica even at high SDI/MFI conditions. Although cake layers were found after DCMD experiments, they do not seem to be related to MD flux decline.
- Due to different operational conditions with pressure-driven membrane filtration system like NF/RO, MD fouling tests using silica and alginate which were used in this study were not correspond to tests based on SDI/MFI.
- Using the same membrane used in MD test, submerged MF experiments were performed and compared with those in the MD operation. The fouling tendency in MD by silica was qualitatively correlated with that in MF. However, the MD fouling by alginate was difficult to be related to the MF fouling.
- Further study will be carried out for quantitative prediction of MD fouling under various conditions by considering the differences in fouling mechanisms between pressure-driven and thermally-driven membrane separation.

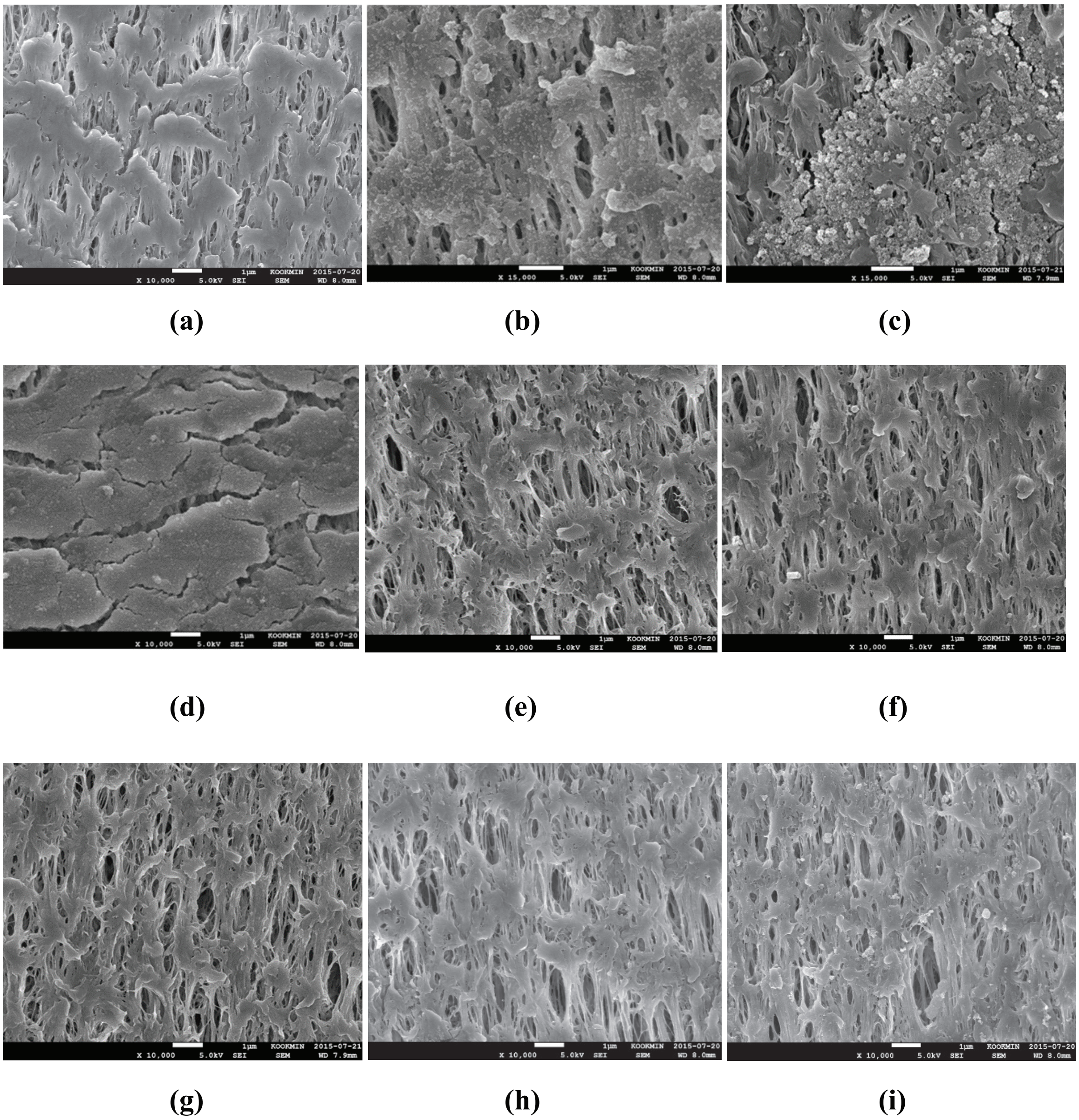


Fig. 7. SEM photograph after DCMD tests: (a) an intact (unfouled) membrane, (b)–(d) silica 50–120 ppm and (e)–(i) alginate 1.8–285 ppm.

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